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A STUDY OF LAMINAR FLOW PHENOMENA UTILIZING A DOUBLY REFRACTING LIQUID

PROGRESS REPORT 1

BY

FRED N. PEEBLES, Assistant Professor
   of Chemical Engineering
JOHN W. PRADOS, Research Assistant
EDWARD H. HONEYCUTT, JR., Research
   Assistant

The Engineering Experiment Station and
The Department of Chemical Engineering
The University of Tennessee
Knoxville 16, Tennessee
July, 1954

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ABSTRACT

Aqueous solutions of a commercial dye known as milling yellow exhibit flow double refraction and, as a result, visual observation of liquid flow phenomena can be made using plane polarized light. This report describes the activities on a research project concerned with the quantitative interpretation of such optical patterns. The long range objectives of the study include the development of an experimental method for the determination of liquid velocity distribution and flow direction based upon the optical behavior of milling yellow solutions. Presently, the work is supported financially by the Office of Naval Research, Contract Nonr-811 (04).

The method of approach to the problem and the experimental methods being employed to measure the optical and viscous properties of milling yellow solutions are described. The optical properties are being determined with a polariscope which includes a concentric cylinder flow cell and the necessary optical components. A concentric cylinder viscometer and a capillary viscometer are being used to make viscosity measurements.

Typical experimental results show that the double refraction of milling yellow solutions is approximately proportional to the velocity gradient. Viscosity measurements indicate that the milling yellow solutions exhibit non-Newtonian behavior in the low velocity gradient range.

A schedule for further experimental activities is presented.

A STUDY OF LAMINAR FLOW PHENOMENA UTILIZING A DOUBLY

REFRACTING LIQUID

Progress Report 1

INTRODUCTION

In 1953 it was found that aqueous solutions of a commercial dye, designated as milling yellow, exhibited flow double refraction, i. e., stresses resulting from the liquid motion bring about light interference patterns of plane polarized light passing through the moving liquid. Such light interference patterns seemed to be reproducible for any arbitrary liquid flow condition. A brief description of this phenomenon as exhibited by milling yellow solutions and the experimental techniques involved was presented by Peebles, Garber, and Jury (8)* at the Third Midwestern Conference on Fluid Mechanics. Photographs and high speed motion pictures of various laminar and turbulent flow examples were shown at that time. Typical light interference patterns are shown in Figure 1.

The preliminary work with the doubly refracting solutions of milling yellow indicated that such solutions could be utilized effectively as a means of liquid flow visualization and even quantitative interpretation of the light interference patterns might be possible. This latter aspect would permit the determination of velocity distribution and flow direction in wholly laminar flow cases and within laminar boundary layers of turbulent flow situations without the use of measuring probes. The attractive features of such an experimental method are obvious. With this long range objective in mind, a research proposal was

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* These numbers refer to the LIST OF REFERENCES, p. 48.

(a) Laminar Flow Through An Orifice
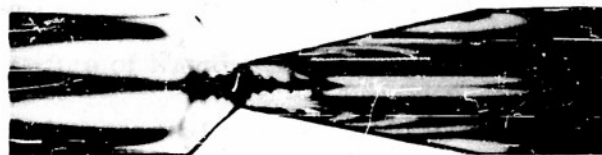
(b) Laminar Flow Through A Nozzle
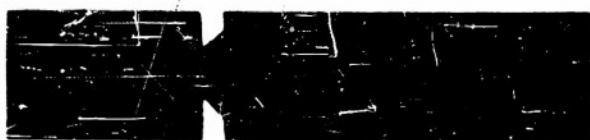
(c) Turbulent Flow Through An Orifice;
Reynolds Number Approximately 5000

(d) Turbulent Flow Through An Orifice;
Reynolds Number Approximately 10,000

(e) Turbulent Flow Through A Nozzle
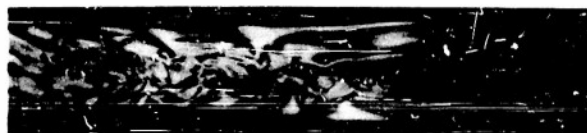
(f) Turbulent Wake Of An Airfoil Section

Figure 1. Typical polarized light interference patterns resulting from flow
double refraction of aqueous milling yellow solutions

submitted by the Engineering Experiment Station and the Department of Chemical Engineering of the University of Tennessee to the Office of Naval Research for financial support in carrying out such a project. This proposal was accepted by the Office of Naval Research and Contract Nonr-811(04) was initiated to support the research program for the period September 1, 1953, to September 30, 1954. Actual notification and authorization of the work was received about November 1, 1953.

The specific objectives embodied in Contract Nonr 811(04) include research in flow visualization along these two directions:

1) To investigate in detail the method of preparing doubly refractive aqueous solutions of milling yellow or other suitable materials, to determine the viscosity and density of such liquids, and to determine the extent of double refraction of milling yellow solutions as a function of velocity gradient.

2) To investigate the quantitative interpretation of simple laminar flow stress patterns using the procedures of photoelastic stress analysis with appropriate modifications.

This report describes the progress on this research project for the period November 1, 1953, to June 30, 1954. It is submitted to the Mechanics Branch of the Office of Naval Research. Additional distribution to other agencies and interested individuals is indicated by the Distribution List.

The report includes an outline of the method of approach toward the long range objectives and a description of experimental methods and preliminary results of the optical properties and the viscosity of milling yellow solutions. In a concluding section of the report a schedule of experimental activities is given.

METHOD OF APPROACH

As indicated in the previous section, it is the long range objective of this research project to make quantitative interpretation of light interference patterns produced in a moving liquid by double refraction of plane polarized light. Such a quantitative interpretation would permit the determination of liquid velocity at various points in the flowing stream without the use of measuring probes inserted into the stream. Perhaps this is an optimistic objective as the basic mechanisms of double refraction in general are ill defined and cannot be described simply. Further, this branch of the field of optics is an unfamiliar one to the engineers who have the need for such a method of measurement. However, engineers have made effective use of double refraction in another well known application, i. e., the method of photoelastic stress analysis. The particular application of double refraction to photoelasticity is quite analogous to the flow double refraction of liquids and it is felt that perhaps the same degree of success can be attained in the analysis of flow stress in moving liquids as in the case of photoelastic stress analysis in solids. In the following paragraphs the basic phenomenon of double refraction is reviewed and the proposed method of application to the objectives listed will be discussed.

A liquid which exhibits the phenomenon of flow double refraction or streaming birefringence behaves optically no differently from other liquids when at rest. It is isotropic with respect to the passage of light through it, and light travels through the liquid at a definite velocity independent of direction. This velocity is inversely proportional to the refractive index of the liquid. When such a doubly refracting liquid at rest is viewed between crossed polarizing prisms or plates, no light is transmitted since the plane polarized light from

the polarizer is unchanged during its passage through the liquid and is extinguished by the analyser.

When a doubly refracting liquid such as a milling yellow solution is set into motion, changes in the optical properties occur. Macroscopically, these changes can be detected in that the velocity of light through the liquid now varies with direction, i. e., the liquid is temporarily anisotropic. Furthermore, it is found that at each point in the moving liquid there is a characteristic direction, called the optic axis, in which light does move at a velocity independent of the direction of propagation. A light wave which vibrates in the plane normal to the optic axis is called the ordinary ray and its velocity is related to the ordinary index of refraction, n_o. Any light wave passing through the liquid which vibrates in a plane parallel to the optic axis is called the extraordinary ray and its velocity is related to the extraordinary index of refraction, n_e.

Effectively, a stressed doubly refracting liquid divides plane polarized light wave vibrations into two components, one parallel to the optic axis and the other perpendicular to the optic axis. Since these two waves move at different velocities proportional to n_o and n_e, one is retarded relative to the other and at various points in the liquid the intensity of the resultant wave is zero owing to destructive addition of the two components. In this case, when the moving doubly refracting liquid is viewed between crossed polarizing plates, a light interference pattern is produced similar to those pictured in Figure 1. Previous investigators have found that the phenomenon can be described quantitatively by the equation

$$n_e - n_o = f\left[\mu \frac{dv}{dr}\right] \tag{1}$$

where

n_e = refractive index of light vibrating parallel to the optic axis

n_o = refractive index of light vibrating normal to the optic axis

f = functional designation

μ = liquid viscosity

$\dfrac{dv}{dr}$ = local velocity gradient

As will be shown in another section of this report, the points of zero light intensity, or the interference fringes, observed by viewing the moving doubly refractive liquid between crossed polarizers are related directly to the refractive index difference, $n_e - n_o$. Hence, if it is desired to analyse the light interference patterns resulting from double refraction of polarized light and thus determine the velocity gradient at various points in the moving liquid, it is necessary to know the functional relation, f, expressed by equation (1) for the particular liquid being employed. In the case of double refraction in solids as applied to the photoelastic stress analysis, it has been assumed that the corresponding functional relation was linear. A similar linear functional relation has been found to apply for pure liquids which exhibit double refraction.

Essentially, an experimental determination of the relationship between the refractive index difference, $n_e - n_o$, and the liquid stress, $\mu \dfrac{dv}{dr}$, for milling yellow solutions will furnish a calibration of the liquid which can be used in the determination of unknown velocity gradients. Before proceeding with the description of the method being employed to determine this calibration, it is appropriate to indicate that it is also possible to determine flow direction from the light interference pattern obtained by viewing a moving double refractive liquid between crossed polarizers.

The determination of flow direction in the moving liquid results from the fact that the angle between the optic axis of the double refractive liquid and and local flow stream line in the moving liquid is, in some cases, a characteristic property of the liquid. (This is the case for flow double refraction in pure liquids.) For milling yellow solutions this angle is a function of the velocity gradient for low values of the velocity gradient and the relationship between these variables must be determined experimentally.

Since it is anticipated to use milling yellow solutions as a medium for the analysis of liquid flow cases, it is essential to know the density and viscosity of these solutions. Previous work and this present work indicate that the determination of the viscosities of milling yellow solutions is a task of significant magnitude. This is borne out in that data on the apparent viscosity of some milling yellow solutions show non-Newtonian behavior at the low velocity gradients. The preliminary viscosity data were measured with a concentric cylinder type of viscometer which permitted measurements only up to velocity gradients of 60 sec.$^{-1}$. A capillary viscometer and the necessary accessory equipment has been set up for the measurement of viscosities of milling yellow solutions at velocity gradients up to 2000 sec^{-1}.

It is felt that at present the scope of activities should be limited to the two general areas described in the preceding paragraphs, i. e., the determination of the basic optical properties and the viscosity of milling yellow solutions. Then, depending on the nature of the experimental results, analytical procedures similar to those employed in photoelastic stress analysis can be devised for the quantitative interpretation of unknown flow double refraction stress patterns.

This type of approach furnishes basic flow information for various solutions of milling yellow from which the solution having the most desirable properties can be selected for the photoviscous flow analysis. Such information perhaps will shed some light on the basic micro-mechanisms which are responsible for the double refraction of milling yellow solutions. The information available suggests that the magnitude of double refraction for these solutions is significantly greater than that reported for any pure liquid in which double refraction results from molecular orientation. This raises many questions concerning the nature of solution of milling yellow dye, that perhaps the milling yellow "solutions" are really suspensions or dispersions of the solute in colloidal form in water. It has been proposed by Jury (5) that the second phase in such a suspension is not a solid but an intermediate "liquid crystal" phase or mesophase. As indicated previously, much of the interpretation of double refraction phenomena in terms of basic mechanisms is conjecture and cannot be described simply at this time. The experimental program outlined will furnish specific information on the aqueous milling yellow system which will permit the answering of some of these questions.

OPTICAL PROPERTIES OF AQUEOUS MILLING YELLOW SOLUTIONS

The optical properties of a liquid which exhibits flow double refraction can be described quantitatively in terms of the difference between the extraordinary and ordinary refractive indices of the liquid, $n_e - n_o$, and the angle between the optic axis of the liquid and the flow streamline. The quantity, $n_e - n_o$, is designated the amount or extent of double refraction, according to Edsall (2). The angle between the optic axis of the double refracting liquid and the flow streamline is usually represented by the symbol χ and is called the extinction angle (2). Both of these quantities are functions of the velocity gradients existing in the doubly refracting liquid. In this section an optical analysis of the double refraction phenomenon is presented in order to explain the method employed in this work for the measurement of double refraction and the extinction angle. The details of the apparatus used and typical experimental data are also given.

Passage of Plane Polarized Light Through a Doubly Refracting Liquid

In order to describe how a doubly refracting liquid produces the optical interference pattern which is characteristic of the particular flow case, consider the following simple case of an element of moving liquid fluid, under constant stress throughout, and illuminated by parallel rays of light through crossed polarizing plates. Schematically this arrangement is shown as Figure 2.

Thus, when plane polarized light vibrating in a direction parallel to OX is transmitted by the polarizer and then passes through the doubly refracting material, it is broken into two plane polarized components. These are indicated in Figure 2 as OQ for the component vibrating parallel to the optic axis and OP for the other component vibrating perpendicular to the optic axis.
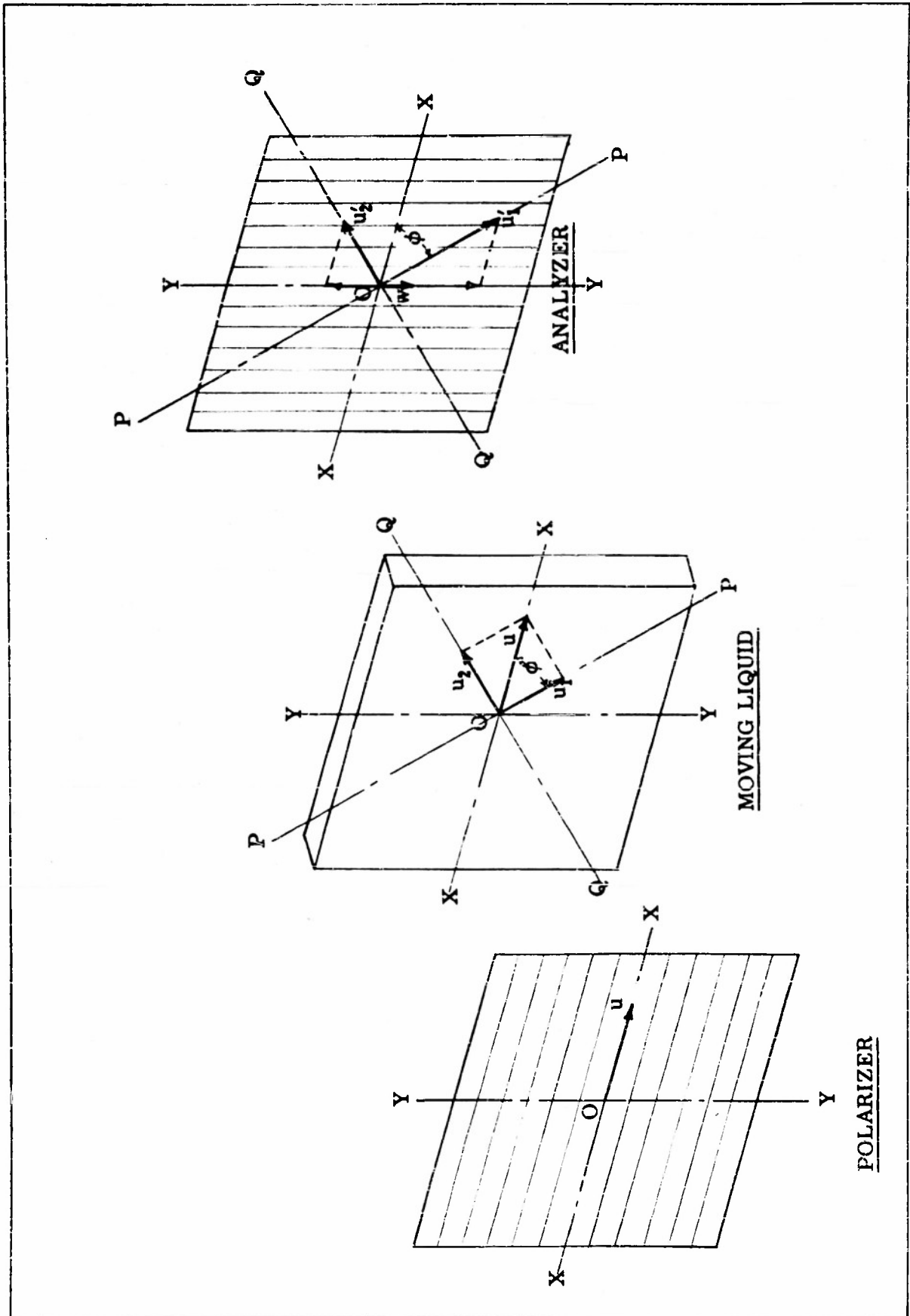
Figure 2. Schematic representation of plane polarized light passing through a doubly refracting liquid

If the equation for periodic motion of the plane polarized light vibration transmitted by the polarizer is expressed as a function of time by

$$u = a \sin 2\pi/\lambda \ (ct - x) \tag{2}$$

where

u   =   displacement of the vibration from the axis O-O at time, t

a   =   amplitude of the vibration

λ =   wave length of the light vibration

x   =   arbitrary phase retardation indicating that the displacement is not necessarily zero at t = 0

c   =   velocity of light in air

then the equations of vibration for the components parallel to OP and OQ respectively for the light entering the doubly refracting liquid are

$$u_1 = a \cos \phi \sin 2\pi/\lambda \ (ct - x) \tag{3}$$
$$u_2 = a \sin \phi \sin 2\pi/\lambda \ (ct - x)$$

where

u_1 =   displacement of the vibration component in the direction OP

u_2 =   displacement of the vibration component in the direction OQ

ϕ =   angle made by OP with the plane of vibration of light from the polarizer (OX)

Since the two components u_1 and u_2 do not travel with the same velocity through the doubly refracting liquid, the slower will emerge some distance, δ, behind the faster. This relative retardation can be expressed quantitatively by the equation of vibration for the light leaving the liquid as

$$u_1' = a \cos \phi \sin 2\pi/\lambda \ (ct - x) \tag{4}$$
$$u_2' = a \sin \phi \sin 2\pi/\lambda \ (ct - x - \delta)$$

where

u_1', u_2' = displacement of the vibration components leaving the doubly

refracting liquid in the direction of OP and OQ respectively

δ = relative retardation produced by the doubly refracting liquid

Now when u_1' and u_2' reach the analyzer, only light vibrating parallel to OY is transmitted. Thus, the vibration equation of the light transmitted through the analyzer can be represented as the resultant wave obtained by adding the components of u_1' and u_2' parallel to OY. This resultant, w, is given by

$$w = u_1' \sin \phi - u_2' \cos \phi \qquad (5)$$

This vector addition of u_1' and u_2' expressed by equation (5) is also shown diagrammatically in Figure 2.

The values of u_1' and u_2' from equation (2) are substituted into equation (5) which, after incorporating the results of basic trigonometric identities, gives the equation of the light vibration transmitted by the analyzer,

$$w = \left[a \sin 2\phi \sin \pi\delta/\lambda\right]\cos (ct - x - \delta/2) \qquad (6)$$

where [a sin 2ϕ sin $\pi\delta/\lambda$] represents the amplitude of the vibration.

Equation (6) indicates the conditions for the existence of the points of light interference, or zero light intensity, in the optical pattern transmitted by the polarizer. Thus, since the intensity of transmitted light is proportional to the square of the amplitude of the vibration, it is noted that the intensity will be zero under two conditions, i. e.,

$$2\phi = N\pi, \phi = N\pi/2 \qquad (7)$$

or,

$$\pi\delta/\lambda = N\pi, \delta = N\lambda \qquad (8)$$

where

$$N \quad = \quad 0, 1, 2, 3, \text{------}$$

Equations (7) and (8) indicate that the light interference pattern produced by a doubly refracting liquid placed between crossed polarizers is composed of various dark regions, or fringes. These fringes exist at points in the liquid where (a) the normal to the optic axis of the liquid is oriented at an angle $\phi = N\pi/2$ with respect to the light transmission axis of the polarizer, or (b) the relative retardation, δ, produced by the passage of plane polarized light through the doubly refracting liquid is a distance equal to an integral number of wavelengths of the incident light.

Measurement of the Extinction Angle, χ

In order to interpret flow double refraction fringe patterns, the angle which the optic axis of the flowing liquid makes with the liquid streamline, of the extinction angle, must be known. Measurements of this angle can be made conveniently in a concentric cylinder type of apparatus described by Edsall (3), which is being used in this present work. In this apparatus the liquid under investigation is placed in the annular space between a fixed inner cylinder and a rotating, glass bottomed outer cylinder, and the entire cell is mounted between crossed polarizing plates. Upon illumination of the annular space between the two cylinders by parallel rays from a monochromatic light source, a double refraction light pattern is obtained characteristic of the flow in the annulus. A variable speed drive for the outer cylinder permits a wide variation of velocity gradients to be used in determining the dependence of the extinction angle on the velocity gradient. Figure 3 shows the assembly of the various components
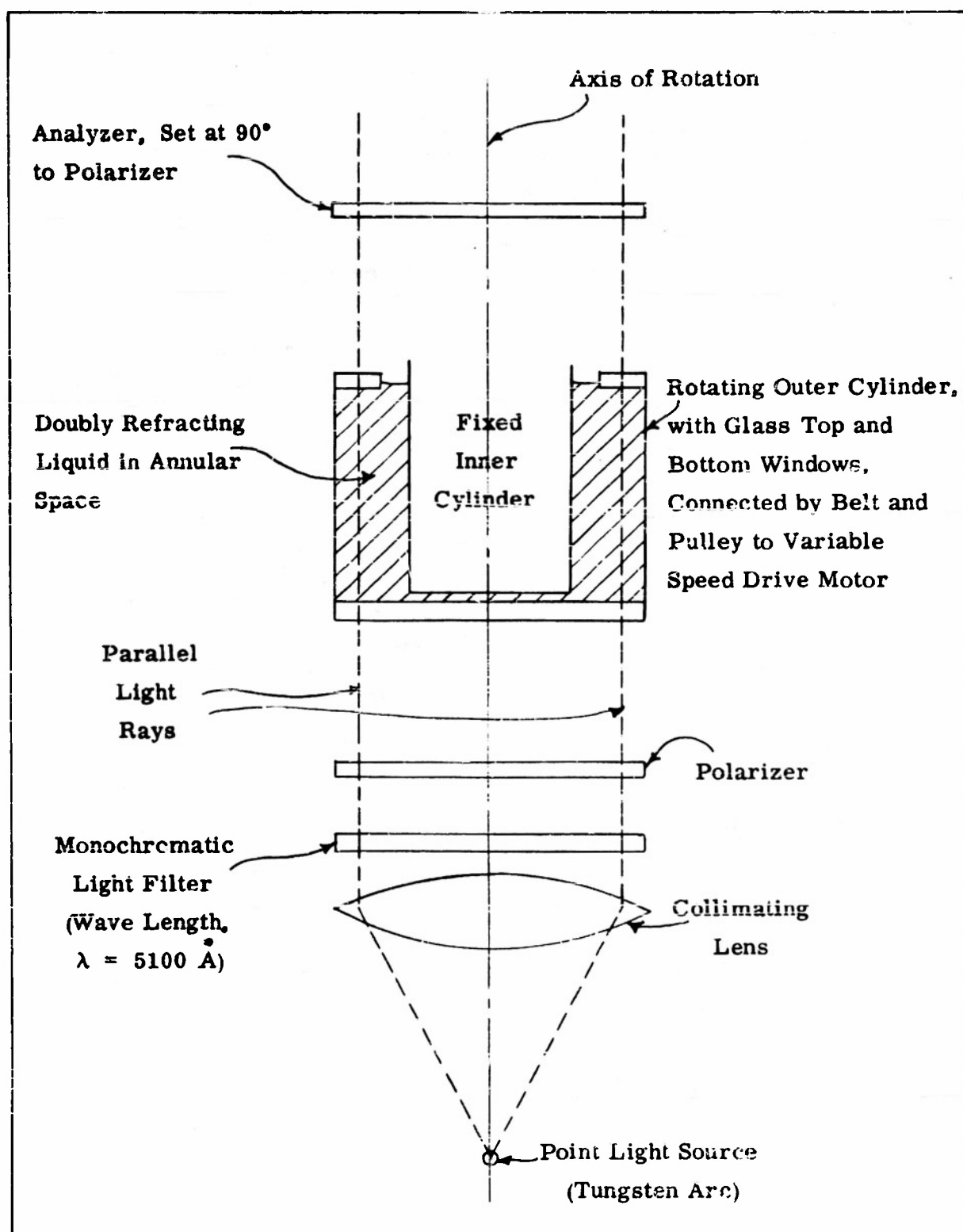
Figure 3. Schematic diagram showing the components
of the concentric cylinder polariscope
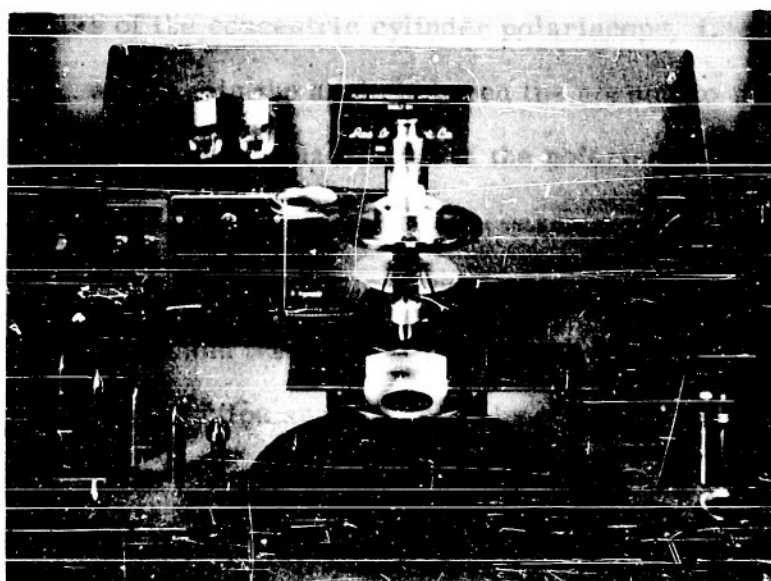
Figure 4. Photographs of the concentric cylinder polariscope

schematically and Figure 4 shows two photographs of the unit. For the remainder of this report this instrument is referred to as the concentric cylinder polariscope.

For laminar flow conditions the light interference pattern produced by doubly refracting solutions of milling yellow in the concentric cylinder polariscope includes four dark areas each separated by 90 degrees extending radially across the annulus. This optical pattern, usually designated as the cross of the isocline, is oriented at some definite position relative to the plane of polarization of the polarizer of the polariscope which is characteristic of the doubly refracting liquid, and may be a function of the particular velocity gradient. This orientation of the cross of the isocline is the basis of the method used to measure the extinction angle. The relation between the position of the cross of the isocline and the optic axis of the liquid can be illustrated by means of Figure 5, which shows the appearance of the cross of the isocline at a typical orientation to the plane of polarization of the polarizer. On the basis of equation (7) there should be four areas of zero light intensity in the annulus of the concentric cylinder polariscope, i. e., at $\phi = 0$, $\pi/2$, π, $3\pi/2$, -----, where ϕ is the angle between the normal to the optic axis of the moving liquid and the plane of polarization of the polarizer. Further, since the streamlines for flow in an annular space are concentric circles, the extinction angle shown as the angle χ in Figure 5, is equal to the smaller acute angle between the arms of the cross and the plane of polarization, OX.

Measurement of the extinction angle is made by aligning a set of 90 degree cross hairs with the arms of the cross of the isocline, and then reading the corresponding angle on a circular scale as indicated by an index on the cross hairs holder.

Measurement of the Amount of Double Refraction

The second basic optical property of aqueous milling yellow solutions that is
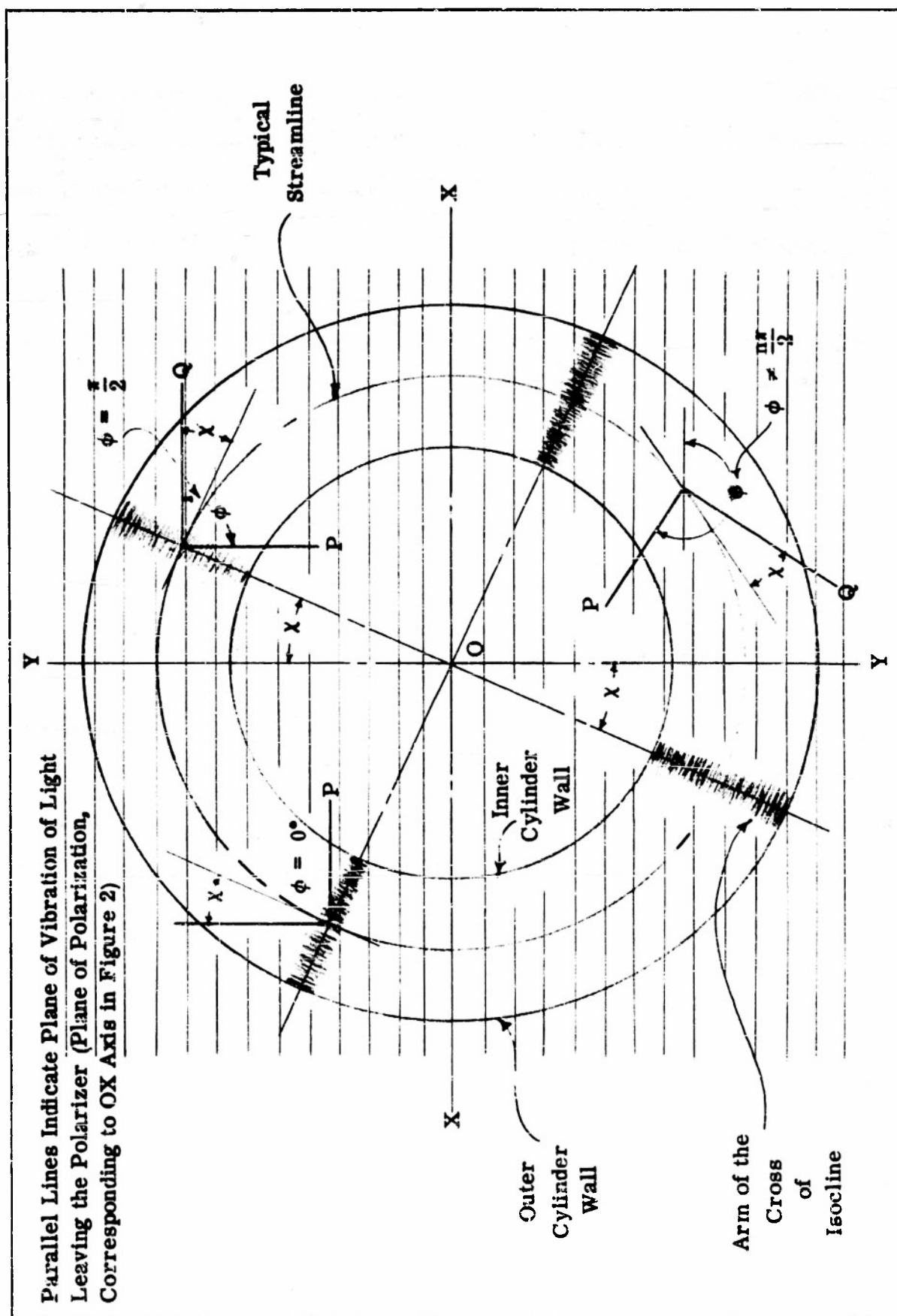
Figure 5. Top view of annulus of the concentric cylinder polariscope showing the cross of isocline and the optical orientation of the doubly refracting liquid

needed is the amount of double refraction, $n_e - n_o$. It was pointed out previously in this report that the quantitative relation between the amount of double refraction and the velocity gradient in the moving liquid must be known in order to pursue interpretation of flow double refraction optical patterns. The concentric cylinder polariscope can also be used to determine the relation between these two quantities for a particular liquid solution. This is done by setting up a known velocity gradient in the liquid, as determined by the speed of the rotating outer cylinder and radial distance between the two cylinders, and then measuring the amount of double refraction. Measurements are made for various velocity gradients by changing the speed of the rotating cylinder.

The amount of double refraction is related to the relative retardation, δ, between the two plane polarized light waves produced by the doubly refracting liquid, and the thickness of liquid through which the light passes. Thus, in the case of the concentric cylinder polariscope used in this study, the depth of liquid in the annular space between the two cylinders constitutes the light path in the doubly refracting liquid. The time required for light to traverse this path will be different for the ordinary ray vibrating normal to the optic axis and the extraordinary ray vibrating parallel to the optic axis. This time difference is given by

$$\Delta t = \frac{S}{c_e} - \frac{S}{c_o} \tag{9}$$

where

Δt = difference in time required for the extraordinary and the ordinary rays to pass through the doubly refracting liquid

S = depth of liquid traversed

c_e = velocity of light vibrating parallel to the optic axis

c_o = velocity of light vibrating normal to the optic axis

This time difference produces the retardation between the two transmitted rays
which is given by

$$\delta = c\Delta t = c \left[\frac{S}{c_e} - \frac{S}{c_o} \right] \tag{10}$$

where

$\qquad$ c $\quad$ = $\quad$ velocity of the transmitted rays in air

Since c/c_e and c/c_o are essentially equal to n_e and n_o,

$$n_e - n_o = \frac{\delta}{S} \tag{11}$$

Equation (11) gives the relation needed to calculate $n_e - n_o$ from a measurement
of δ for a particular liquid depth, S.

The measurement of δ can be made by two methods using the concentric
cylinder polariscope. The first method has its basis in the application of
equation (8), which indicates that, when δ is an integral number of wave lengths
of the light being employed, a dark area or fringe should exist in the doubly re-
fracting liquid as observed in the polariscope. Thus, when monochromatic
light is passed through the doubly refracting liquid in the concentric cylinder
polariscope with the outer cylinder at rest, the annulus appears dark. This
corresponds to the zero order fringe, N = 0 in equation (8), and indicates zero
retardation. As the outer cylinder is put into motion, the annulus becomes
illuminated and the cross of the isocline is observed, and as the speed of the
outer cylinder is gradually increased, a dark circle appears in the annulus,
superimposed on the cross of the isocline at some definite cylinder speed. This
corresponds to the first order fringe, N = 1 in equation (8), and the retardation
is equal to λ. The speed of the outer cylinder is measured at this condition,
and from this speed and the gap thickness of the annulus, the velocity gradient

can be calculated. As the speed of the outer cylinder is continuously increased, the higher order fringes appear consecutively, corresponding to $N = 2, 3, ----$ in equation (8). At the cylinder speeds where each of the fringes appear in the annulus, the speed of the cylinder is measured and the velocity gradient calculated. Then, knowing the wavelength of the light and the liquid depth used, equation (11) can be used to compute the amount of double refraction for each fringe order observed. By measurements of this type, the relation $n_e - n_o$ and velocity gradient for the doubly refracting liquid is determined.

For the measurement of fractional orders of retardation, the method just described is not suitable and an optical compensator must be used. Generally, measurement of double refraction by a compensator involves inserting into the optical system a known thickness of a doubly refracting material in order to compensate for the amount of double refraction being measured. In this present work a polaroid quarter wave plate is used as a compensator. It is inserted into the optical system of the concentric cylinder polariscope between the liquid cell and the analyzer with the optic axis of the quarter wave plate parallel to the plane of polarization of the polarizer. Under these conditions an additional $\lambda/4$ retardation is added between the two plane polarized light waves emerging from the doubly refracting liquid in the cell. The insertion of the quarter wave plate does not affect the appearance of the cross of the isocline as long as the position of the analyzer is not changed. However, upon rotation of the analyzer, the arms of the isocline cross are observed to move toward each other in a scissor-like manner until finally they collapse into only two dark areas displaced 45 degrees from the position of the cross before turning the analyzer. The angle, Θ, which the analyzer is turned to achieve this condition is related to the retardation pro-
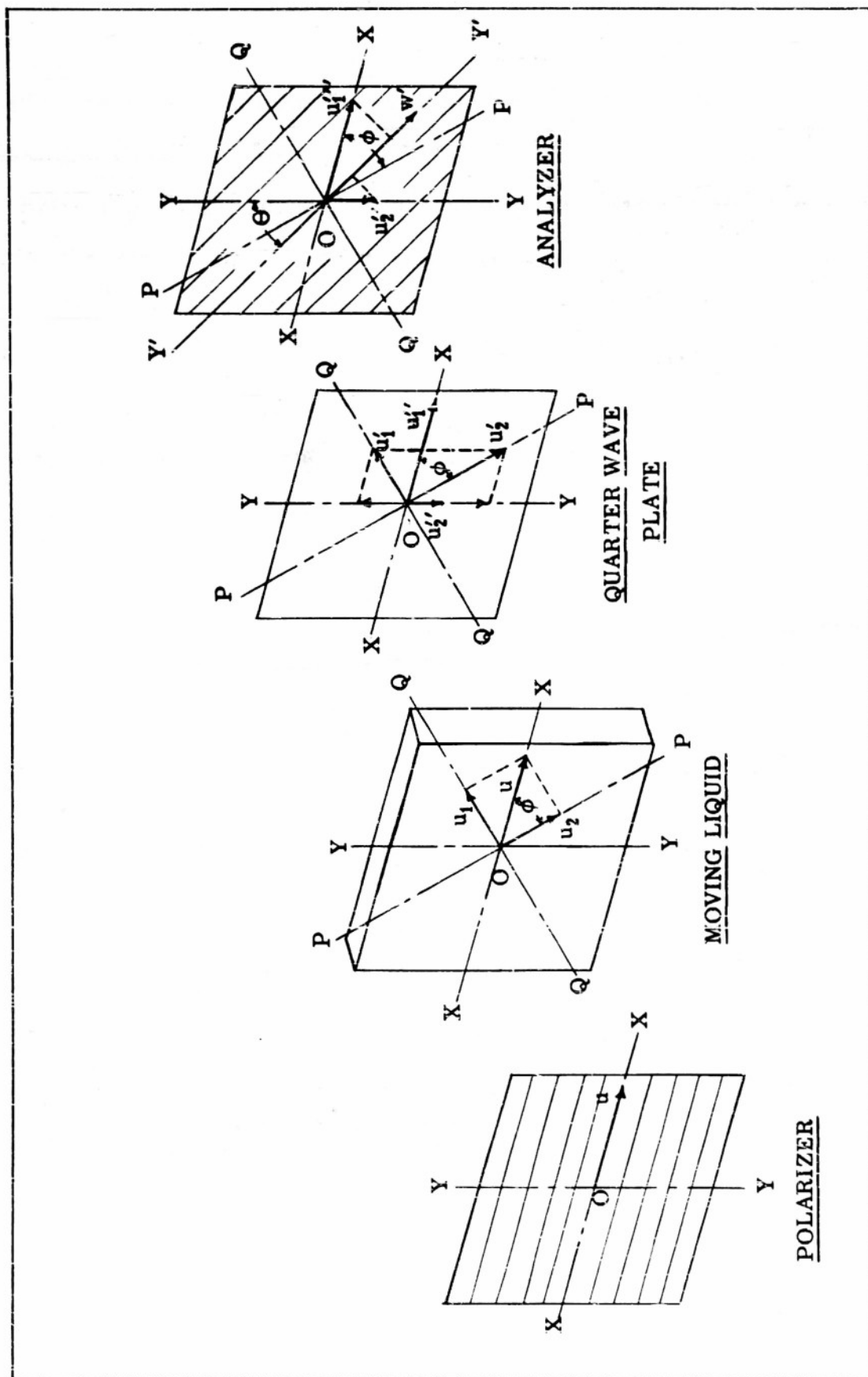
Figure 6. Schematic representation of polarized light passing through a doubly refracting liquid with a quarter wave plate inserted between the liquid and the analyzer

duced by the doubly refracting liquid. This relation can be shown by means of the following analysis.

Referring to Figure 6, u_1', and u_2' represent as before the two light waves leaving the doubly refracting liquid which now enter the quarter wave plate. The quarter wave plate transmits two light waves vibrating in the directions parallel and normal to the optic axis of the quarter wave plate, which are OX and OY respectively. Thus the components of u_1' and u_2' in the direction of OX and OY as they enter the quarter wave plate are

$$u_1'' = u_1' \cos \phi + u_2' \sin \phi \qquad (12)$$

$$u_2'' = u_1' \sin \phi - u_2' \cos \phi$$

where

u_1'' = displacement of the vibration component entering the quarter wave plate in the direction OX

u_2'' = displacement of the vibration component entering the quarter wave plate in the direction OY

Substituting the values of u_1' and u_2' from equation (4) into equation (12),

$$u_1'' = a \cos^2 \phi \sin 2\pi/\lambda \, (ct - x) + a \sin^2 \phi \sin 2\pi/\lambda \, (ct - x - \delta) \qquad (13)$$

$$u_2'' = a \sin \phi \cos \phi \sin 2\pi/\lambda \, (ct - x) - a \sin \phi \cos \phi \sin 2\pi/\lambda \, (ct - x - \delta)$$

As u_1'' and u_2'' pass through the quarter wave plate, one is retarded with respect to the other by an additional retardation of $\lambda/4$. Representing the displacements of these two waves emerging from the quarter wave plate as u_1'' and u_2'', they are given by

$$u_1'' = a \cos^2 \phi \sin 2\pi/\lambda \, (ct - x) + a \sin^2 \phi \sin 2\pi/\lambda \, (ct - x - \delta) \qquad (14)$$

$$u_2'' = a \sin \phi \cos \phi \cos 2\pi/\lambda(ct-x) - a \sin \phi \cos \phi \cos 2\pi/\lambda \, (ct - x - \delta)$$

The transformation of u_2'' to u_2'' results owing to the trigonometric relation

$$\sin (\alpha - \pi/2) = \cos \alpha \qquad (15)$$

where

α = any angle measured in radians

When u_1'' and u_2'' reach the analyzer only the component vibrating parallel to OY is transmitted; i. e., u_1'' is extinguished completely and u_2'' passes through the analyzer unchanged. This light wave transmitted by the analyzer is really the same as that without the quarter wave plate inserted, since it can be shown that

$$u_2'' = - [a \sin 2\phi \sin \pi\delta/\lambda] \sin 2\pi/\lambda (ct - x - \delta/2) \tag{16}$$

The amplitude, and hence the intensity, is numerically equal to that given by equation (6) and is subject to the same conditions for zero intensity, $\phi = N\pi/2$ and $\delta = N\lambda$, (N = 0, 1, 2, 3, ----). Therefore, with the quarter wave plate inserted into the light path of the concentric cylinder polariscope and the analyzer in the original orientation, the four arms of the cross of the isocline and the occurence of the various fringe orders are observed unchanged.

For the measurement of the retardation the analyzer is rotated clockwise to some angle, Θ, where the cross of the isocline collapses into two dark areas separated from each other by 180 degrees. At this point the plane of polarization of the analyzer lies along OY' as shown in Figure 6. In this position the vibration equation of the light transmitted by the analyzer is

$$w' = u_1'' \sin \Theta + u_2'' \cos \Theta \tag{17}$$

where

w' = displacement of the vibration component transmitted by the analyzer in the direction OY'

Or substituting the values of u_1'' and u_2'' from equation (14) into equation (17),

$$w' = a \sin \Theta [\cos^2 \phi \sin 2\pi/\lambda (ct - x) + \sin^2 \phi \sin 2\pi/\lambda (ct - x - \delta)]$$
$$+ a \cos \Theta \sin \phi \cos \phi [\cos 2\pi/\lambda (ct - x) - \cos 2\pi/\lambda (ct - x - \delta)] \tag{18}$$

Now the two dark areas of the pattern transmitted by the analyzer occur at points in the liquid displaced $\pi/4$ radians from the arms of the cross of the isocline, or at $\phi = \pi/4$ and $5\pi/4$. For these values of the angle, ϕ,

$$\sin\phi\cos\phi = \sin^2\phi = \cos^2\phi = 1/2 \tag{19}$$

Using the information expressed by equation (19) and other trigonometric identities, it can be shown that

$$w' = a\sin(\Theta - \pi\delta/\lambda)\sin 2\pi/\lambda(ct - x - \delta/2) \tag{20}$$

Since at these two points in the rotating liquid the intensity of the light transmitted by the analyzer is zero, this means that the amplitude of w' is zero, which in turn indicates that

$$(\Theta - \pi\delta/\lambda) = N\pi \tag{21}$$

For any value of $N = 0, 1, 2, 3, ---$, equation (21) indicates that the retardation is directly proportional to the angle Θ, and the wave length of the light used and is given by

$$\delta = \frac{\Theta\lambda}{\pi} \tag{22}$$

It can be shown that for the rotation of the analyzer in the counter-clockwise direction equation (22) also applies and that the two dark areas resulting from the collapse of the cross of the isocline are positioned at $\phi = 3\pi/4$ and $7\pi/4$.

For the concentric cylinder polariscope being used in this work, measurement of the angle Θ is limited to $\Theta \leqq \pi$; or fractional orders of retardation can be measured up to and including the first order with the quarter wave plate compensator. For retardations greater than the first order, the first method of measuring double refraction can be used. It is recalled that this method is limited to the measurement of only integral orders of retardation. For both methods of measurement described, equation (11) is used to calculate the extent of double refraction from the experimental values of δ and S.

Experimental

The methods for measuring the amount of double refraction and the extinction angle which were described in the preceding sections have been used for milling yellow solutions, and seem to give satisfactory results. Typical results are shown in Figures 7, 8, 9, and 10. For the results shown, an inner cylinder r_1 = 0.460 inches and an outer cylinder r_2 = 0.500 inches were used. The velocity gradients were evaluated at the center of the annulus using these dimensions and and angular velocity of the outer cylinder by means of the well known relationship for the laminar flow velocity distribution cited by Edsall (2).

Figure 7 shows the extinction angle for a 1.30 per cent by weight milling yellow solution as a function of the velocity gradient, and Figure 8 gives the corresponding information for a 1.35 per cent solution. It is noted that for both of these solutions, the extinction angle approaches a value of 45 degrees for low velocity gradients, and then the extinction angle decreases with increasing velocity gradients to a value near 20 degrees for the 1.30 per cent solution and near 17.5 degrees for the 1.35 per cent solution.

Figures 9 and 10 show the extinction angle and the amount of double refraction of 1.30 and 1.35 per cent milling yellow solutions for velocity gradients up to 300 sec.$^{-1}$. Here it is noted that the double refraction of milling yellow solution is of the order of 10^{-5}. This represents a very large double refraction in comparison to the values for pure liquids, which are in the range of 10^{-10} to 10^{-12}, depending on the velocity gradients. The double refraction found for the milling yellow solutions is comparable to a V_2O_5 colloidal solution investigated by Freundlich et al (4), for which double refractions of approximately 10^{-4} were found. In a classic work by von Muralt and Edsall (7), the double refraction of

rabbit myosin solutions was found to be approximately 10^{-5}, which is considerably smaller than that of the milling yellow solutions. On the basis of these preliminary data and the comparisons cited, it is indicated that the double refraction of milling yellow is not due to molecular phenomena.

The experimental data given in Figures 7-10 were determined for temperatures over the range 27-30°C. It is felt that the scatter of the extinction angle results from this variation in the temperature, as the concentric cylinder polariscope was designed to be operated in a constant temperature laboratory. The necessary modifications are being made to thermostat the laboratory in which this double refraction work is being conducted, and this should improve the precision of the double refraction measurements.
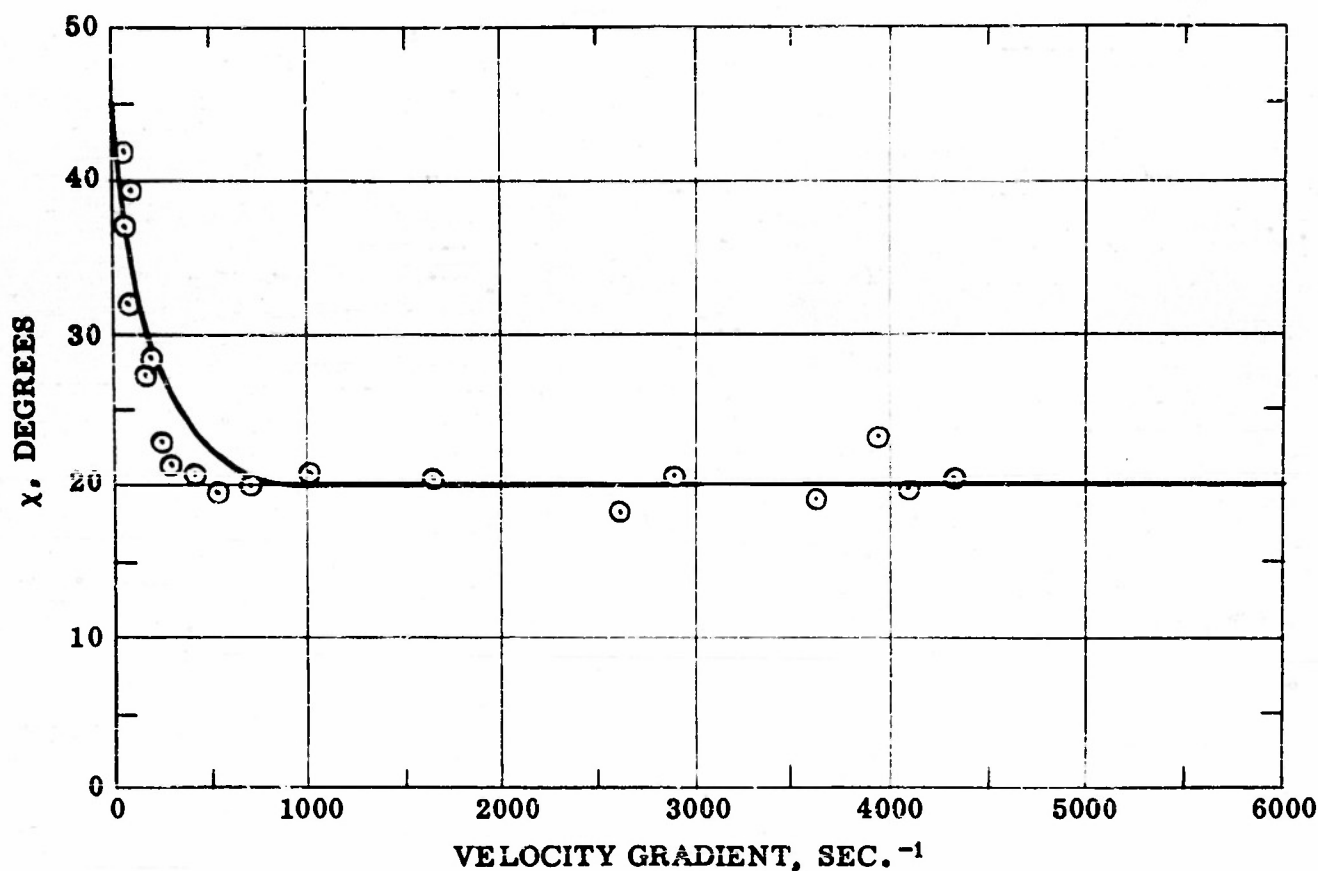
Figure 7. Extinction angle as a function of the velocity gradient for a
1.30% milling yellow solution at high velocity gradients
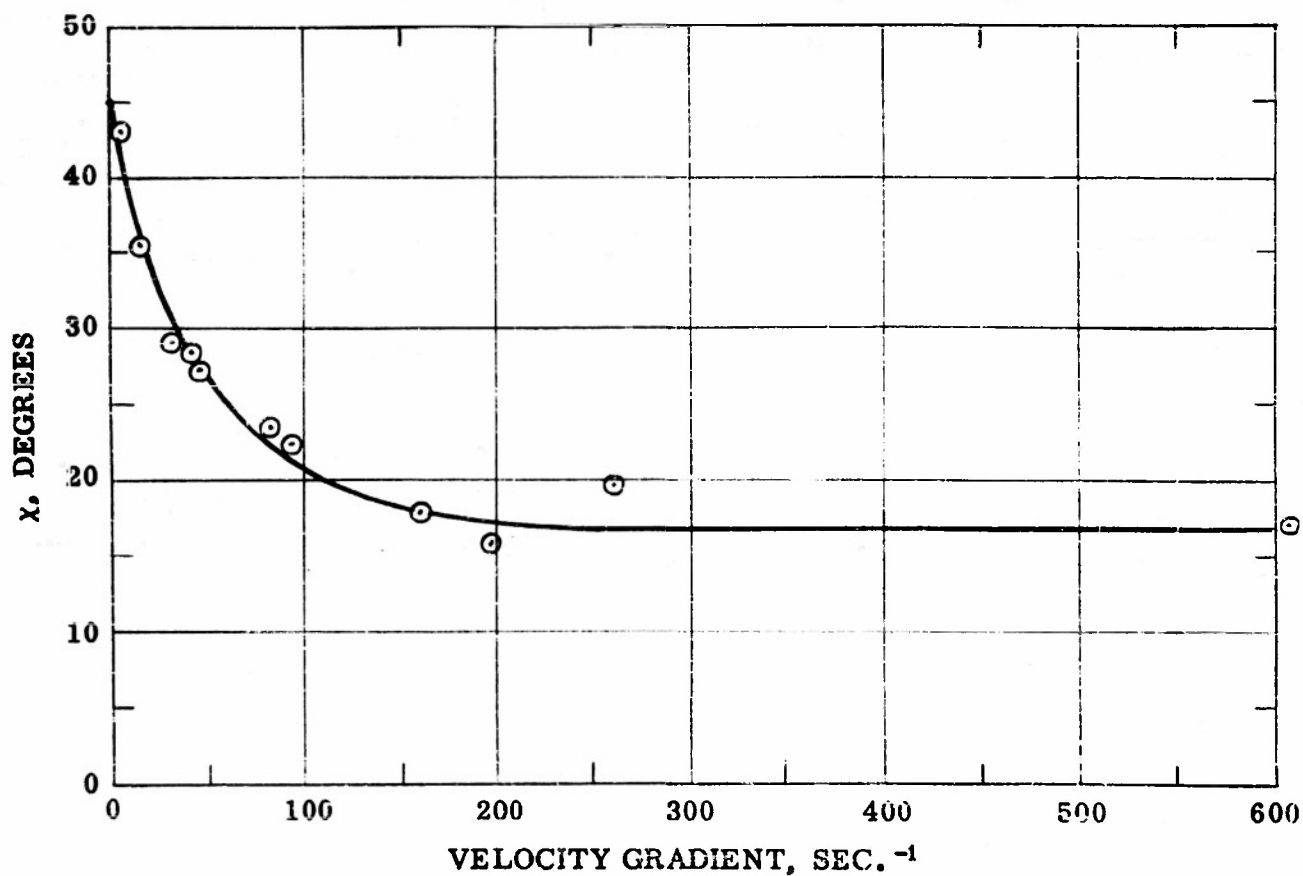

Figure 8. Extinction angle as a function of the velocity gradient for a
1.35% milling yellow solution at high velocity gradients
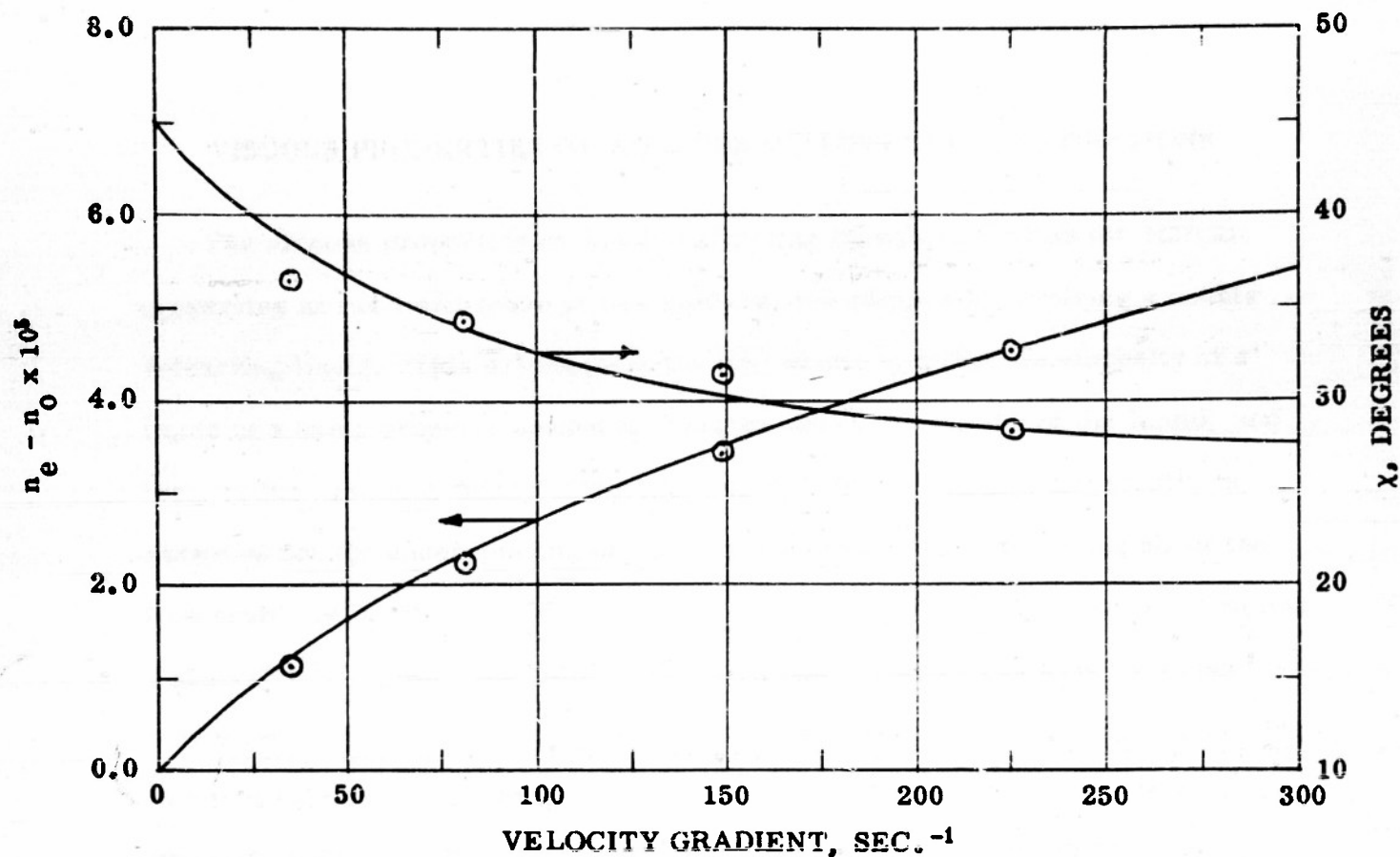
Figure 9. Double refraction and the extinction angle as a function of velocity gradient for a 1.30% milling yellow solution
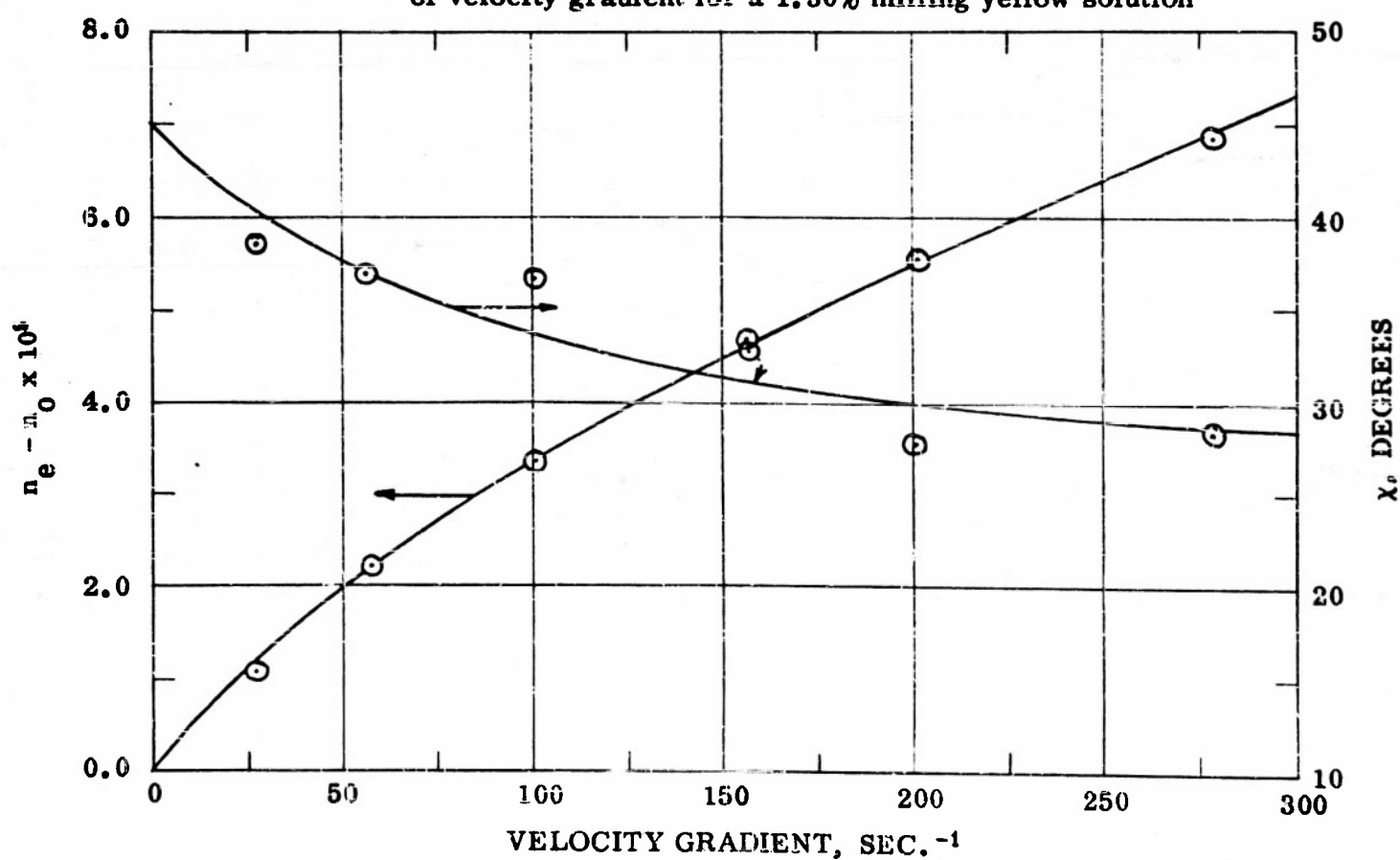
Figure 10. Double refraction and the extinction angle as a function of velocity gradient for a 1.35% milling yellow solution

VISCOUS PROPERTIES OF AQUEOUS MILLING YELLOW SOLUTIONS

The viscous properties of doubly refracting liquids as well as the optical properties are of importance in this study of flow phenomena utilizing a doubly refracting liquid. This arises from the well known fact that the viscosity of a liquid is a basic property needed to describe the flow behavior of the liquid, and that, in the case of a doubly refracting liquid, information on the viscosity is essential for the understanding of the micro-mechanisms which bring about the flow double refraction. That the viscous and optical behavior of doubly refracting liquids are inter-related is well known and many investigators have reported information on both properties of various doubly refracting liquids. An outstanding example is the work of Robinson (9) who made simultaneous double refraction and viscosity measurements with a concentric cylinder type of flow cell.

The nature of viscous resistance of liquids is well known and the literature on viscosity is voluminous giving viscosity data on various fluids, experimental methods and techniques, and theoretical considerations. A review of the more recent literature on viscosity of liquids has been completed and will be presented in a future progress report.

In the preliminary work on the behavior of milling yellow solutions, limited viscosity data as determined by means of a rolling ball viscometer were reported (8). With the initiation of this present study it was decided that a viscometer of a type that would permit viscosity determination for well defined velocity gradients was needed in order to obtain absolute data, rather than relative viscosities which perhaps were characteristic of the viscometer being used. The rolling ball viscometer is such a relative viscosity instrument and the use of this instrument was eliminated in favor of a concentric cylinder viscometer.

The merits of this latter type of viscometer for the determination of absolute vis-
cosities have been established by many investigators.

The viscosities of four aqueous milling yellow solutions were determined at
25°C, 30°C, and 35°C for shear rates up to 60 sec.$^{-1}$ with a MacMichael concentric
cylinder viscometer. In the following sections, details of this experimental work
are described and typical results are presented. A discussion of the results and
plans for additional experimental work is also given.

The MacMichael Concentric Cylinder Viscometer

As indicated, the MacMichael viscometer is of the concentric cylinder type
and is widely used in industrial practice for both control work and fundamental
measurements. In essence it consists of a rotatable turntable, to which a
cylindrical cup is attached, and a stationary cylinder suspended from a fixed
support by a wire. When liquid is placed in the annular space formed by the cup
and the suspended stationary cylinder, and the turntable is rotated, viscous
shear in the liquid produces a torque on the stationary cylinder, thus twisting
the cylinder about the axis of the supporting wire. At equilibrium the torque in
the liquid is balanced by the resisting torque of the twisted wire and the inner
cylinder is moved from the zero flow position by some definite angular dis-
placement. This angular displacement, as indicated by an index pointer on a
circular scale, is related to the viscosity of the liquid and the angular speed of
the rotating cup. For laminar flow conditions in the annulus and negligible end
effects, this relation is

$$\theta_t = \frac{r_i^2\, L_w\, S\, \mu}{4 E_s\, r_w^4} \left.\frac{dv}{dr}\right|_{r_i} \tag{23}$$

where

Θ_t = angular displacement of the inner cylinder

r_i = radius of the inner cylinder

L_w = length of supporting wire

S = depth of liquid in the annulus

$\bar{E}_s$ = modulus of elasticity in shear of the supporting wire

r_w = radius of the supporting wire

μ = liquid viscosity

$\left.\dfrac{dv}{dr}\right|_{r_i}$ = radial velocity gradient at the inner cylinder

For a particular instrument with a specific wire used to support the inner cylinder, equation (24) can be written as

$$\Theta_t = K\mu\Omega \tag{24}$$

where

K = an instrument constant

μ = liquid viscosity

Ω = angular velocity of the outer cylinder

Equation (24) was used to relate the angular displacement of the stationary inner cylinder of the MacMichael viscometer to the viscosity of the liquid and speed of the turntable.

For all measurements, an outer cup, r_2 equal to 0.592 inches, was used with an inner cylinder, $r_1 = 0.542$ inches. This gave an annulus of 0.050 inches thick, and for the speeds of the turntable available, velocity gradients up to 60 sec.$^{-1}$ could be attained. Wires of various sizes were used with this one cylinder arrangement in order to obtain measureable response over a wide viscosity range.

Figure 11 shows schematically an assembly of the MacMichael viscometer as used, and Figure 12 shows two photographs of the unit.

Experimental

Viscosity determinations were made for aqueous solutions of milling yellow containing 1.31, 1.47, 1.51, and 1.77 per cent milling yellow by weight at 25°C, 30°C, and 35°C. No precise means of thermostating the solution in the annulus of the viscometer was available, and a water bath adjusted to the test temperature was used to maintain the temperature within ± 0.5°C of the desired value.

This method of temperature was adequate in making measurements on Bureau of Standards petroleum oils for calibration purposes. Figure 13 shows a typical set of calibration data. Here the angular displacement of the inner cylinder measured in MacMichael degrees (based on an arbitrary circular scale containing 300 units) is plotted versus the turntable speed in revolutions per minute. On the basis of equation (24), these plots should be linear for a Newtonian liquid whose viscosity is independent of the rate of shear. This is supported by the experimental data, and plots of this type were used to evaluate the instrument constant for each wire used in the viscometer at each of the test temperatures.

Figure 14 shows a typical plot of the angular displacement of the inner cylinder versus turntable speed for the 1.47 per cent milling yellow solution at the three temperatures. Here it is noted that a non-linear curve is obtained at 25°C and 30°C. This indicates that this solution is non-Newtonian, i. e., viscosity is a function of the shear rate at these temperatures and at the shear rates used in the tests. At 35°C the non-Newtonian behavior is not exhibited by this solution. Except for the 1.77 per cent solution, this behavior is representative of the doubly refracting solutions of milling yellow tested to date.

1. Inner cylinder
2. Outer cylinder
3. Supporting wire
4. Angular displacement scale
5. Water bath
6. Turntable
7. Variable speed
   motor housing
8. Leveling screw

115 V A. C.
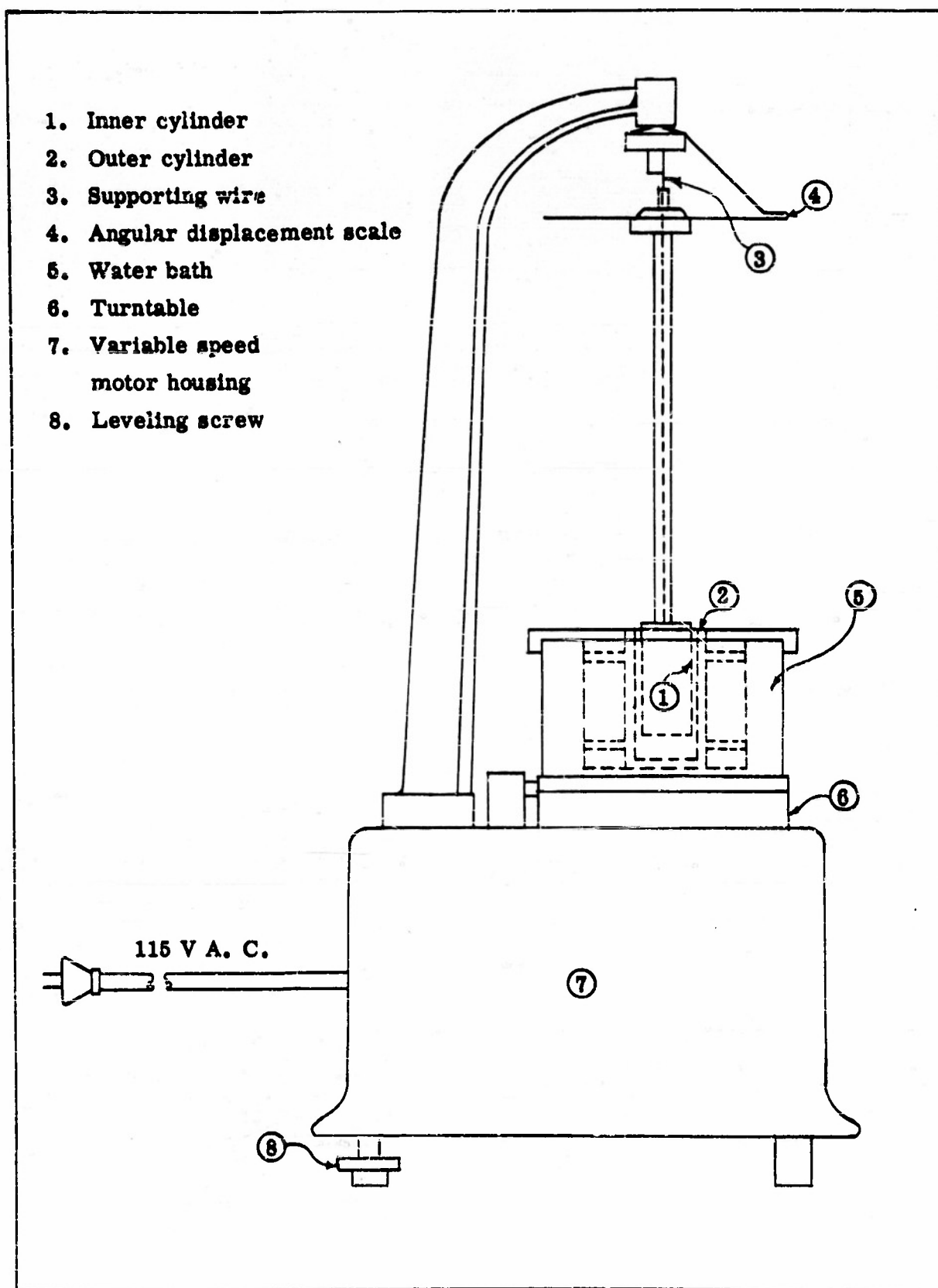
Figure 11. Schematic diagram of the MacMichael viscometer
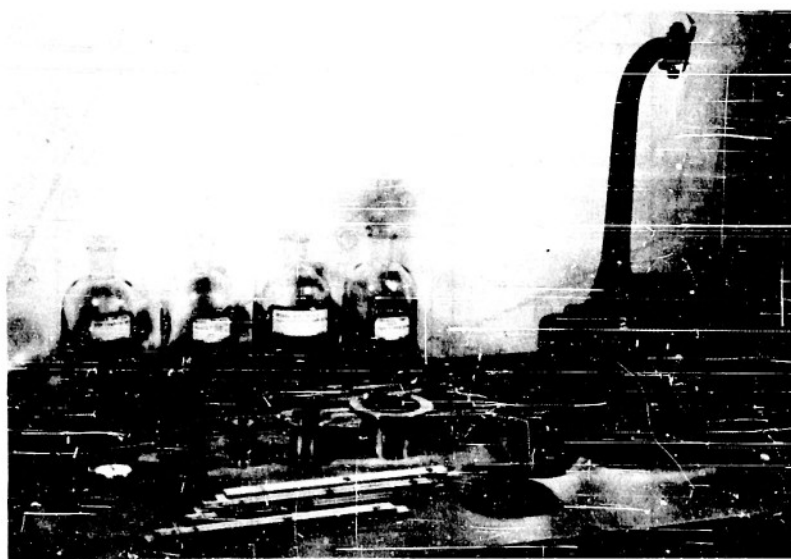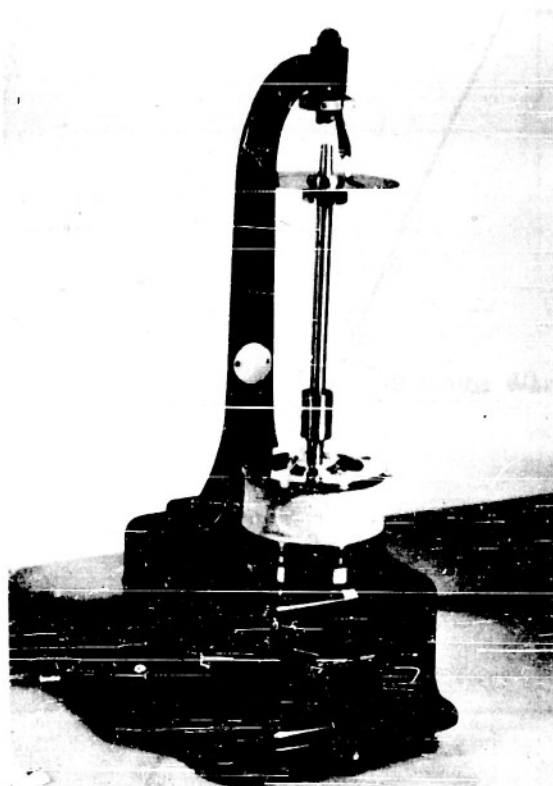
Figure 12. Photographs of the MacMichael viscometer
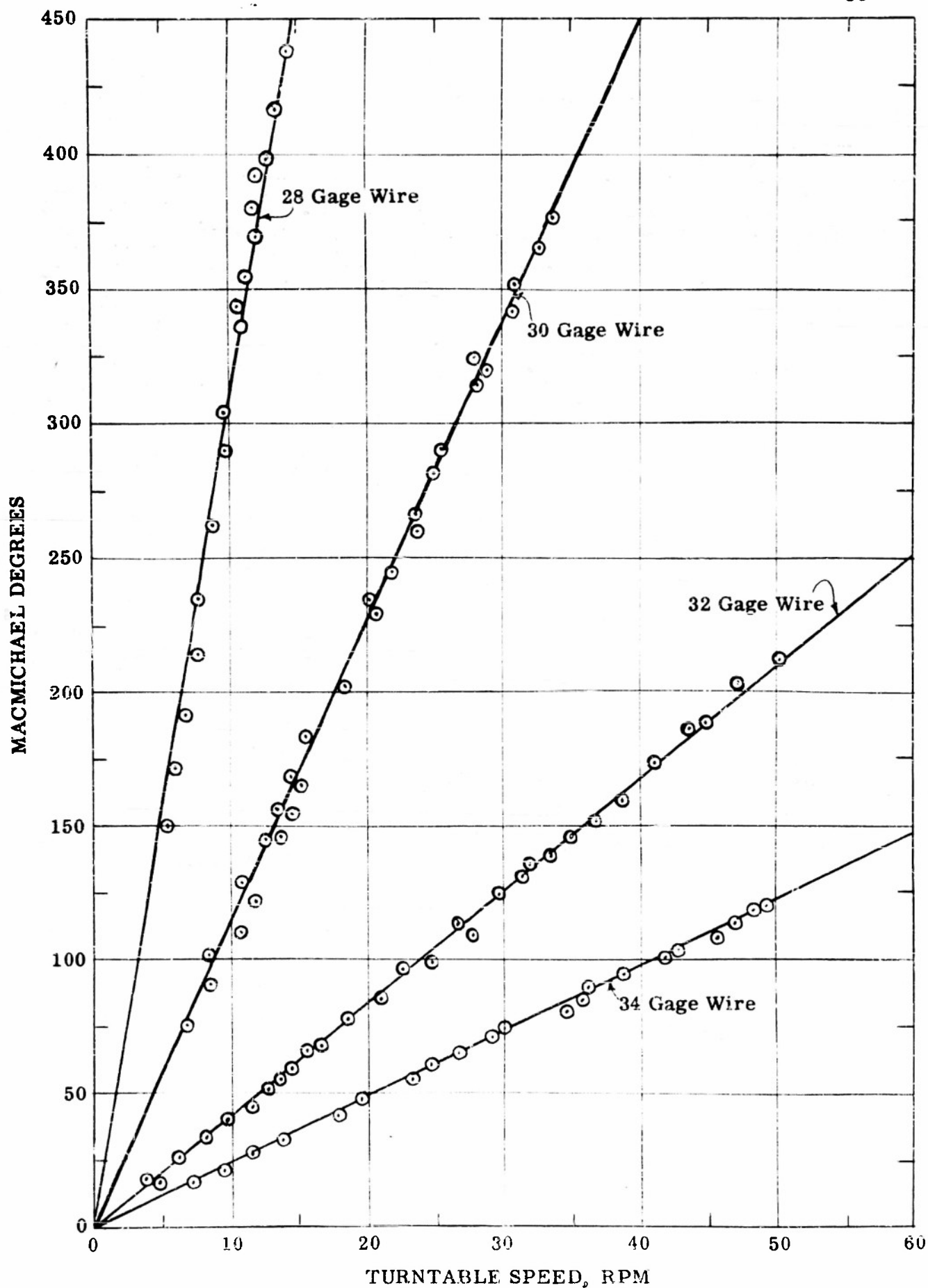
Figure 13. Typical calibration data obtained with the MacMichael Viscometer using a Bureau of Standards petroleum oil
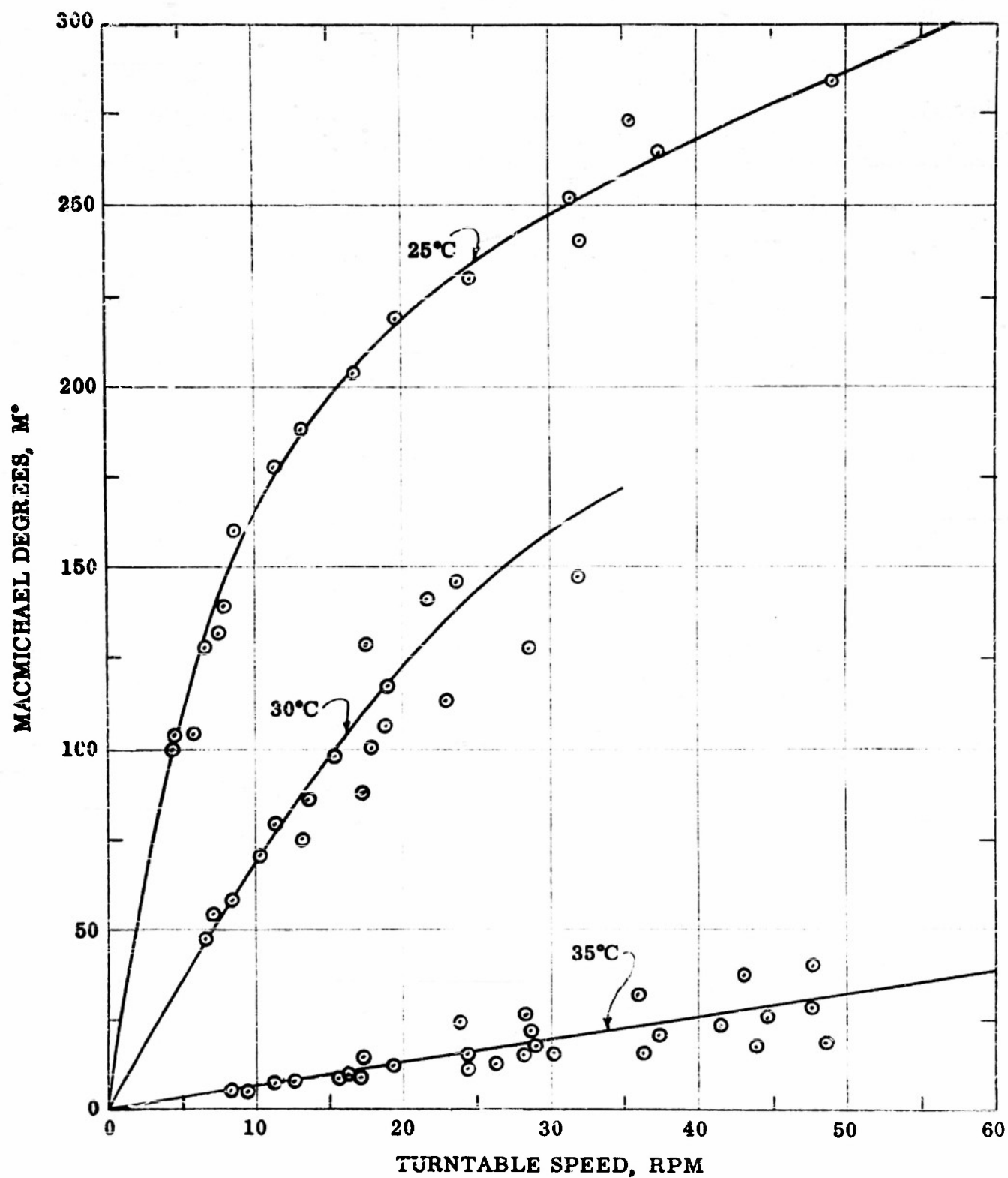
Figure 14. Angular displacement versus turntable speed for the 1.47% milling yellow solution

Figure 15 shows the apparent viscosities as a function of the shear rate, or velocity gradient, for the 1.47 per cent milling yellow solution. Here, the apparent viscosity, μ_a , is calculated from equation (24) which is based upon the assumption of a constant viscosity independent of velocity gradient. Since this assumption is not necessarily valid for the milling yellow solutions, the viscosity is designated as an apparent viscosity. It is noted that for the 25°C and 30°C runs the apparent viscosity is quite high at the very low velocity gradients and decreases sharply with increasing velocity gradient. Further, in the very low velocity gradient range, the apparent viscosity is markedly temperature dependent, exhibiting a 30 fold increase over the range 35°C to 25°C. It is felt that this marked temperature dependency caused the variation in the data points, since with the water bath arrangement it was possible to maintain temperature conditions only within ± 0.5°C.

The Capillary Viscometer

The viscosity measurements obtained with the MacMichael viscometer indicate clearly that some of the doubly refracting solutions of milling yellow are non-Newtonian in the velocity gradient range of zero to 60 sec.$^{-1}$ and that the variation in viscosity with temperature in this velocity gradient range is quite pronounced. On the basis of these results, it was decided to change the method of measurement to permit better control of the liquid temperature under the test conditions and to obtain viscosity data over a greater range of velocity gradients.

From the review of the literature on experimental methods of measuring the viscosity of non-Newtonian liquids, it seemed that a capillary viscometer described by Bingham (1) satisfied the two requirements specified. This type of viscometer involves measuring the time required for a definite volume of the
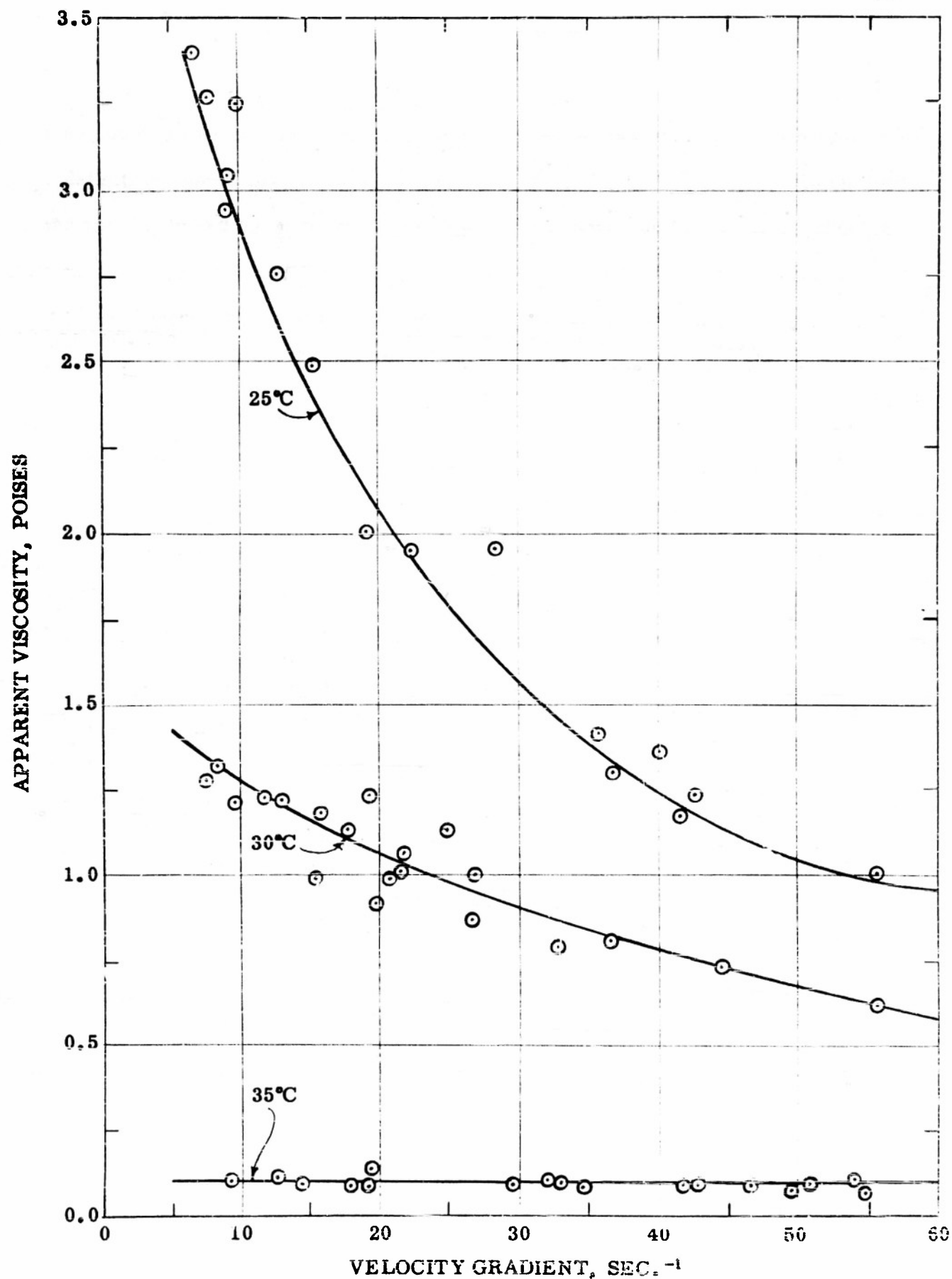
Figure 15. Apparent viscosity as a function of velocity gradient for a 1.47% milling yellow solution

test liquid to flow through a glass capillary tube of known diameter and length under a definite pressure drop across the capillary tube. Such a capillary tube containing the test liquid can be submersed in a thermostated water bath and, thus, precise temperature control can be obtained. Further, by employing capillary tubes of various diameters and using various pressures to force the liquid through the capillary tube, a wide range of velocity gradients can be obtained.

Figure 16 pictures an experimental facility involving a capillary viscometer which has just been assembled. As indicated in Figure 17, which shows a schematic diagram of this equipment, compressed nitrogen is used to blow the liquid through the capillary tube. The pressure drop across the capillary tube is measured with either of two manometers; one a mercury manometer, the other a water manometer. The range of pressure drop, which can be measured with the manometers, permits the determination of the liquid viscosity at velocity gradients up to 2000 sec.$^{-1}$.

Non-Newtonian Flow in a Capillary Viscometer

Many investigators have pointed out the use of the capillary viscometer is not suitable to determine the viscosity-rate of shear relationship for non-Newtonian liquids in that the velocity gradient varies across the tube in an unknown manner. An apparent viscosity can be computed from the usual experimental measurements using the familiar Poiseuille formula

$$\mu_a = \frac{\pi R^4 \Delta P}{8 L Q} \tag{25}$$

where

R = radius of the capillary

ΔP = pressure drop across the capillary

L = length of the capillary

Q = volumetric flow rate through the capillary
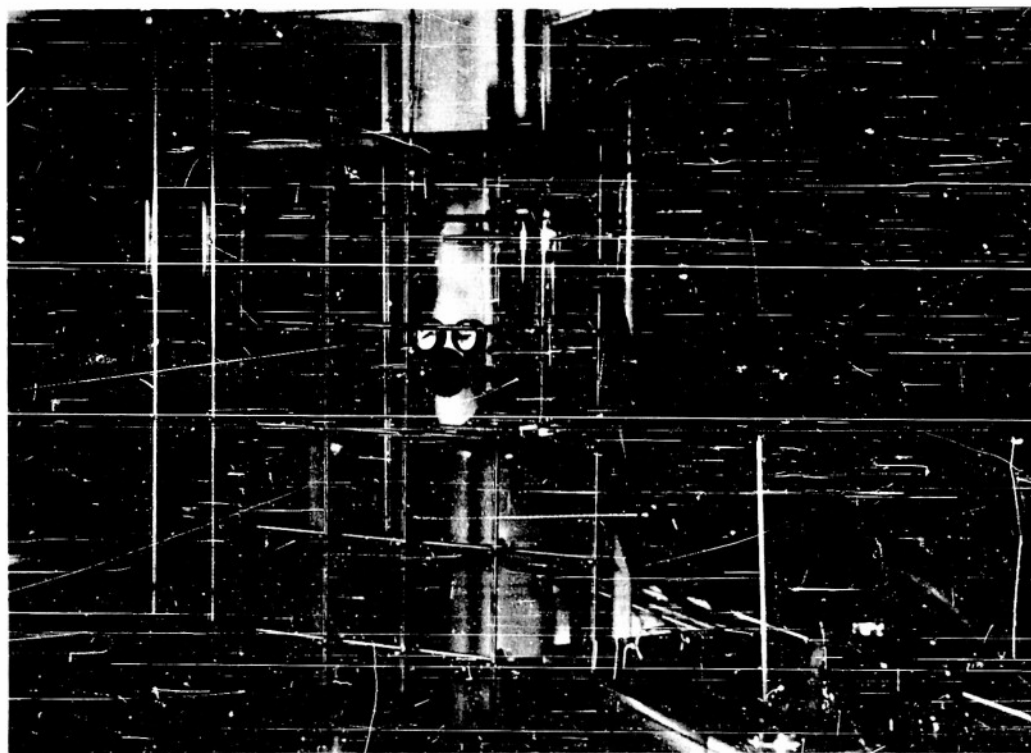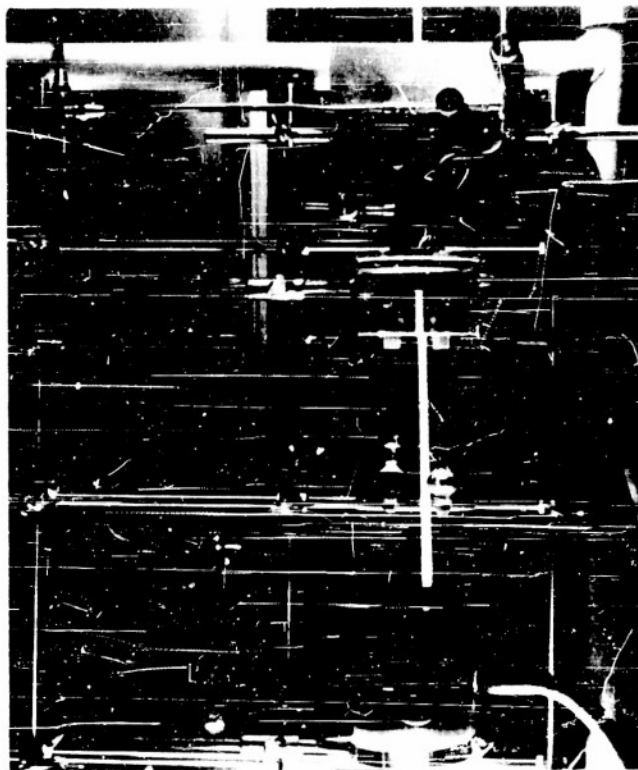
Figure 16. Photographs of the capillary viscometer facility
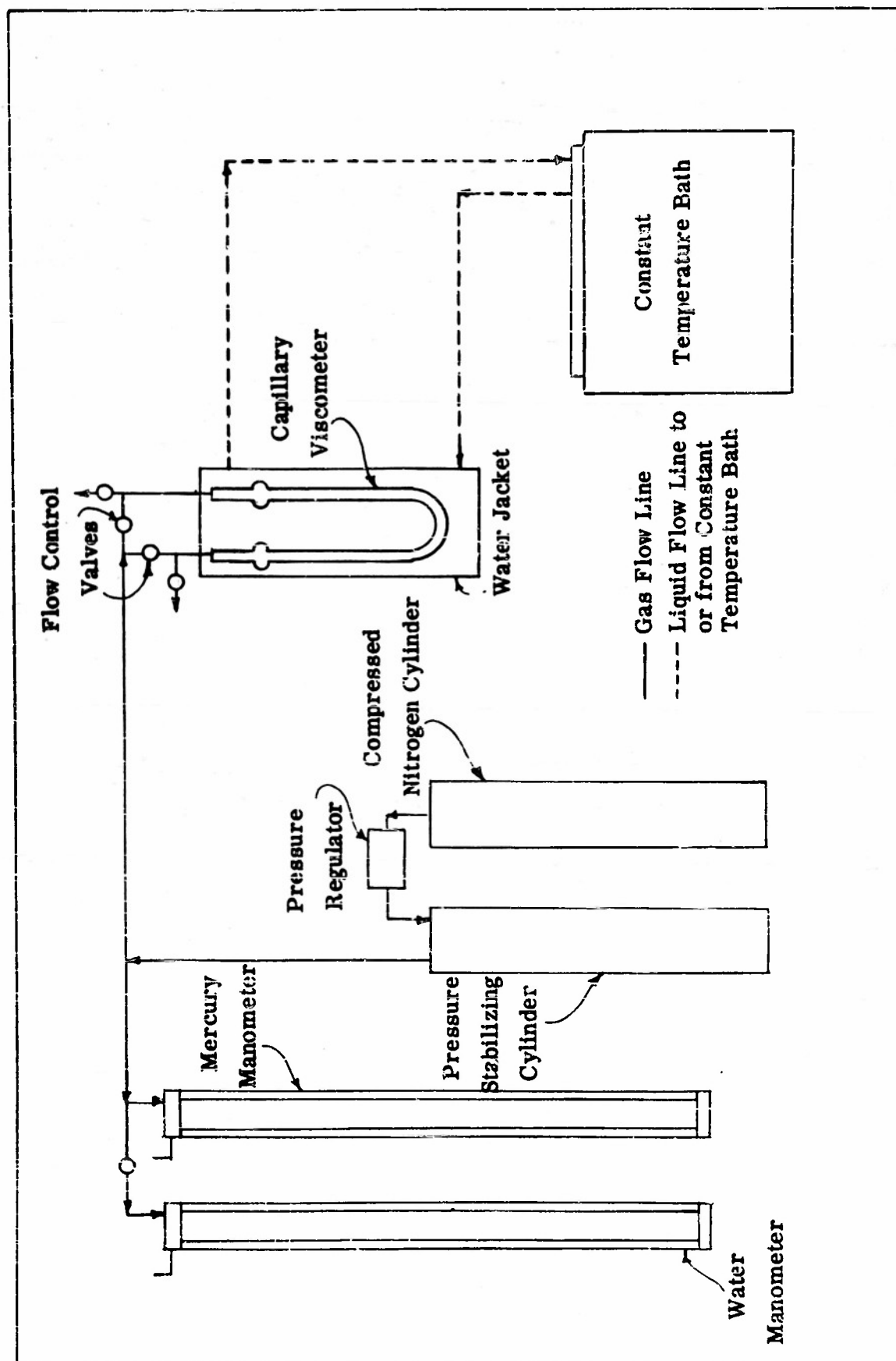
Figure 17. Schematic diagram of the capillary viscometer facility

This apparent viscosity is not equal to the true viscosity defined for flow in a circular tube by

$$\tau = -\mu \, dv/dr \tag{26}$$

where

τ =   local shear stress

dv/dr =   radial velocity gradient

except when the viscosity is independent of the velocity gradient.

Recently Kreiger and Elrod (6) suggested a method for relating the apparent viscosity of non-Newtonian liquids, as determined with capillary or concentric cylinder viscometers, to the true viscosity. Their method, with some modifications, is presented below to show the method of obtaining the true viscosity-rate of shear relationship from experimental measurements of the apparent viscosity of milling yellow solutions. The analysis, as presented, is limited to laminar flow in a circular tube encountered in the capillary viscometer.

Since non-Newtonian flow in a circular tube involves an unknown velocity distribution across the tube radius which is some function of the shear stress, let

$$-dv/dr = g(\tau) \tag{27}$$

where

$g(\tau)$ =   some unknown shear stress function characteristic of the
             non-Newtonian liquid

This unknown function can be related to the apparent viscosity by using equation (27) to compute the total flow in the circular cross-section. Thus,

$$Q = \int_R^0 \pi r^2 \, dv = \int_0^R \pi r^2 \, g(\tau) \, dr \tag{28}$$

The shear stress is a linear function of the radial position in the tube, r, and the pressure drop across the tube length, L

$$\tau = \frac{\Delta P}{2L} r \tag{29}$$

Changing the variable in equation (28) to τ through the use of equation (29),

$$Q = \frac{8\pi L^3}{(\Delta P)^3} \int_0^{\tau_w} \tau^2 \, g(\tau) \, d\tau \tag{30}$$

where

$$\tau_w \quad = \quad \text{wall shear stress at } r = R$$

Now combining equations (25) and (30),

$$\frac{1}{\mu_a} = \frac{4}{\tau_w^4} \int_0^{\tau_w} \tau^2 \, g(\tau) \, d\tau \tag{31}$$

Thus the apparent viscosity of the liquid is a single-valued function of the wall shear stress, τ_w, consequently the function $g(\tau)$ can be obtained by differentiating μ_a with respect to τ_w. Or,

$$g(\tau) = \frac{\tau_w}{\mu_a} - \frac{1}{4} \left(\frac{\tau_w}{\mu_a} \right)^2 \frac{d\mu_a}{d\tau_w} \tag{32}$$

Equation (32) gives $g(\tau)$, which is numerically equal to the velocity gradient, as a function of τ_w and μ_a.

In the use of the capillary viscometer the experimental variables are t, the time necessary for a fixed volume of liquid to flow through the capillary, and ΔP, the pressure drop across the capillary. Further, the experimental data necessary to define the non-Newtonian flow of the liquid involve measuring t for a wide range of ΔP values. Equation (32) may be expressed in terms of these

experimental variables by incorporating the definitions of τ_w and μ_a; or,

$$g(\tau_w) = -\frac{dv}{dr} = \frac{\tau_w}{\mu_a}\left[1 - \frac{1}{4t}\frac{d(\Delta P \cdot t)}{d(\Delta P)}\right] \qquad (33)$$

Combining equations (26) and (32), the true viscosity of the non-Newtonian liquid is

$$\mu = \frac{\mu_a}{\left[1 - \frac{1}{4t}\frac{d(\Delta P \cdot t)}{d(\Delta P)}\right]} \qquad (34)$$

These last two equations are to be used to compute the true viscosity-rate of shear relationship for milling yellow solutions from experimental data obtained with the capillary viscometer. No experimental results from such measurements are available at this time.

SCHEDULE OF EXPERIMENTAL ACTIVITIES

The furtherance of the experimental program described in the previous sections is planned as follows:

1) **Measurement of the extinction angle and the amount of double refraction of milling yellow solutions.** The experimental methods described in this report for these measurements are satisfactory and yield accurate, reproducible results. The quarter wave compensator used to measure fractional orders of double refraction retardation is somewhat limited in application to milling yellow solutions. However, a Babinet-Soliel compensator which will permit measurement of fractional orders of retardation up to six orders has been purchased and is to be delivered by August 1, 1954. It is anticipated that this instrument will have a measuring range adequate for all the milling yellow solutions to be studied.

It was found that the double refraction of milling yellow solutions is temperature dependent. In order to make measurements under controlled temperature conditions, it has been necessary to thermostat the laboratory being used for this work. The necessary air cooling equipment was received recently and now the laboratory temperature can be controlled within $\pm$ 0.3° C of a specified temperature.

It is anticipated that the measurements on the extinction angle and the double refraction of milling yellow solutions can be completed by January 1, 1955.

2) **Measurement of the viscosity of milling yellow solutions.** Three different experimental methods have been employed to make viscosity measurements on milling yellow solutions. The third method based upon the use of the capillary viscometer promises to be the most reliable and accurate of the methods used. Owing to the non-Newtonian characteristics of some milling yellow solutions,

the experimental determination of the apparent viscosity as a function of the pressure drop across the capillary viscometer and the computation of the true viscosity from these measurements involves many experimental observations and a moderate amount of computational time to treat the data. The completion of this work is scheduled for March, 1955.

3) Photoviscous flow analysis of laminar flow phenomena. Upon completion of the double refraction measurements which essentially serve as a calibration of the photoviscous effects of milling yellow solutions, it is planned to attempt a quantitative flow analysis of simple laminar flow cases such as two dimensional flow in a rectangular tube, flow past a cylinder, flow in a converging channel, etc. This work will require a flow double refraction polariscope similar to the type used in the photoelastic stress analysis of transparent solids. There has been no activity on this phase of the long range program and such activity awaits the completion of the double refraction measurements and the acquisition of appropriate polariscope equipment.

This schedule of experimental activities projects beyond the present date of termination of the Office of Naval Research task order authorizing this work. A proposal for extension of the task order for another one year period is being submitted to this agency in the immediate future.

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APPENDIX A

LITERATURE SURVEY ON FLOW DOUBLE REFRACTION

A survey has been made of the literature pertaining to flow double refraction. The results are presented in the following sections, broken down into two major parts. (1) A classification of references under several general headings, each concerning a specific aspect of work in the field. These contain a listing, by number, of the pertinent references and a brief discussion of them. (2) An alphabetical listing of references by authors, giving also the headings and sub-headings under which each reference is classified.

Most of the work considered here deals directly with flow double refraction. However, certain selected references from closely related fields have been included to provide background information. Those articles which include reviews of the field of flow double refraction have been listed under a separate heading for convenience in location. It will be noted that a number of references fall under more than one heading, and are so listed.

Method of Classification

The reference material is grouped under the following headings and sub-headings.

I. Theoretical Material

    A. Initial Observation

    B. Quantitative Theories of Flow Double Refraction

II. Experimental Measurements

    A. Pure Liquids

    B. Polymers

C. Colloids

D. Proteins

E. Other Substances

III. Fluid Flow Analysis with Doubly Refracting Liquids

IV. Equipment and Procedures

V. Background Information

A. General Optical Theory

B. Double Refraction

C. Flow in Concentric Cylinders

D. Relation of Molecular Form to Viscosity and Flow Behavior

E. Electrical and Magnetic Double Refraction

F. Dielectric Measurements

VI. Reviews

I. Theoretical Material

A. Initial Observations: References 3, 41, 89, 90, 97, 140, 149, 150, 151, 154, 162, 186, 212, 260, 276, 277, 278. In the course of some optical experiments in 1866, J. Clerk Maxwell observed the phenomenon of flow double refraction in a viscous liquid (Canada Balsam), stirred by a glass spatula between crossed polarizing prisms. He reported this in 1873 (151) together with some qualitative theories as to its cause; in the same year, Mach (150) reported similar observations. In 1881 Kundt (140) repeated Maxwell's experiments on a number of liquids and Lieck (149) in 1886 studied the double refraction of gelatin. The first quantitative measurements were made in 1888 by de Metz (154) who measured the amount of double refraction in various liquids, and observed a relation between it and the liquid viscosity. Other observations in this period were made by Umlauf (259), Schwedoff (212), Almy (3), Hüil (97), and Reiger (186), but none of these could advance a quantitative theory as to the cause of the phenomenon. A suggestion by Schwedoff, based on his observations, was used by Natanson (162) in developing an elaborate theory of viscosity which was never generally accepted. In 1904 and 1905, Zakrzewski (276, 277), in studies on colloidon, noticed that the extinction angle was not always 45° but decreased as he increased the stress on the liquid. From his results with this and other systems, he concluded that the extinction angle is 45° in pure liquids, but may differ markedly from this value in substances of a colloidal nature. The closest approach to a quantitative theory of flow double refraction in this early period was made by Havelock (89, 90) in 1908, who developed expressions for the amount of double refraction from considerations of the molecular properties of the fluids, but did not derive a relationship between this and the velocity gradient.

B. <u>Quantitative</u> <u>Theories</u> <u>of</u> <u>Flow</u> <u>Double</u> <u>Refraction:</u> References 4, 17, 20, 21, 23, 29-33, 35-41, 50, 51, 66-71, 72, 73, 79, 85, 89, 91-94, 96, 101, 108, 109, 117, 125, 127, 130-139, 142-144, 148, 153, 159-161, 166, 172, 173, 177-179, 181, 183, 185, 187, 188, 190, 193-197, 202, 203, 206, 208, 213, 215, 216, 218, 222, 230, 234, 237, 247-249, 251, 274, 279, 280. In 1915 Freundlich, Zocher, and their collaborators (46, 47, 66-70, 279-280) began extensive studies on vanadium pentoxide sols which were already known to consist of long rod-like micelles which increased in length on standing. From the results of these investigations they concluded that the double refraction in the sols was the result of the orientation of the particles by forces arising from liquid flow. Frey (71) further subdivided the effect into (1) that caused by orientation of normally isotropic particles whose refractive index differs from that of the surrounding fluid. (This "form" double refraction does not exist, therefore, if the solvent has the same refractive index as the particles); (2) that due to anisotropy of the particles themselves. This idea is based on a theory developed by Wiener (272) for the non-flow case, and has been experimentally demonstrated by Lauffer (142-144) in studies on tobacco mosaic virus.

In 1924 and 1925 Vorlander and Walter (265, 266) measured the double refraction of a large number of pure organic liquids. Their results indicated a somewhat different mechanism of double refraction than that for the colloids. Using these measurements to check their work, Raman and Krishnan (183) in 1928 developed the first quantitative theory relating double refraction, molecular properties, and the velocity gradient in the flowing fluid. They assumed the orientation of rigid ellipsoidal molecules to be the cause of the double refraction, and their expressions agreed well with the data of Vorlander and

Walter. However, their equations failed to predict the behavior cf colloidal sols, and solutions of very large molecules. Further, Sadron (193, 194) has cast doubt as to the accuracy of the results of Vorlander and Walter, since some of their measurements were taken in a range where turbulent flow would occur in their apparatus. At present no quantitative theory has been developed for the turbulent region. In 1932 Boeder (20, 21) developed a second quantitative theory based on the simplified case of orientation of unidimensional particles. His work represents a limiting case encountered experimentally—systems of long rigid rod shaped particles—and data on these agree well with his expressions.

During the next 10 years Sadron (193-197, 199), Signer (213, 215, 216, 218, 222), and W. Kuhn (130-133) all advanced quantitative theories which gave good results on a number of systems. W. Kuhn, in conjunction with H. Kuhn, has continued to develop refinements on his original theories up to the present (134-139). In 1939 Peterlin and Stuart (177, 178), after a critical survey of existing theories, derived expressions of a more general nature than those previously advanced. Peterlin (174, 175) and Stuart and Peterlin (234) have also modified and expanded their theories in recent years.

Several investigators have developed theories of flow double refraction within the last 10 years, as the result of investigations into the nature of polymers. Some of the more extensive are the work of Cerf (29-33, 35-40), Joly (108, 109), and Riseman and co-workers (74, 249, 251). Most of the modern theories, while assuming various models for the particles, have considered flow double refraction the result of a combination of two effects: (1) deformation of the particle by forces present in the flow, (2) orientation

of the particle due to the fact that its ends lie in fluid moving at different velocities. In systems of elastic particles the first effect predominates; in systems of long rigid particles the second effect is responsible. In systems of coiled polymer molecules both effects may play an important role. Some very informative measurements have been made by W. Kuhn, Künzle, and Katchalsky (122, 138) on polymer molecules having electrical charges distributed along them. By varying the pH of the solution, they have been able to induce a coiling or uncoiling of the molecular chain owing to varying attraction and repulson between the charges, and a corresponding variation in flow double refraction properties results.

Two excellent reviews have appeared in recent years covering theory and experiment in the field of flow double refraction. A review by Edsall (50) covers work prior to 1942, and one by Cerf and Scheraga covers work from 1942 to 1952 (40).

II. Experimental Measurements

A. Pure Liquids: References 1, 27, 127, 161, 194, 196, 254, 262, 263, 265, 266, 273. The double refraction of a large number of pure organic liquids has been measured by various investigators. In one respect, these substances are ideally suited for fluid flow studies, since their extinction directions make a constant angle (45°) to the streamlines, and most of them show a linear variation of the amount of double refraction with velocity gradient. However, this amount of double refraction is very small except in liquids of extremely high viscosity; and none of the liquids tested so far, whose viscosity is low enough to make them very useful in fluid flow experiments, would show any effect at all in the usual type of flow test equipment.

Double refraction measurements on pure liquids were reported by
Kreuger (127) in 1924, and by Vorlander and his collaborators (262, 263,
265, 266) from 1924 to 1932. Vorlander's measurements covered a very
large number of doubly refracting and non-doubly refracting liquids. He
found the effects to be most pronounced in those liquids whose molecules
were assymetric, and zero for the more symmetric molecules. However,
in 1939 Buchheim, Stuart, and Menz (27) developed an apparatus for measur-
ing extremely small effects, with which they studied benzene derivatives; and
in 1941 Winkler and Kast (273) measured the flow double refraction of benzene
itself. They found a sharp increase in the double refraction when the temper-
ature was decreased to within about 10°C of the freezing point. Further work
on benzene at low temperatures was done by Tsvetkov and Kibardina (254) in
1948.

In 1935 Alcock and Sadron (1) actually used a pure liquid (sesame oil)
in a fluid flow study. The high viscosity of the fluid limited the range of
their results. Sadron (194, 197) also performed tests on pure liquids to
support his contentions as to the inaccuracies in the Raman and Krishnan
theory. In 1952 Narasimhamurty (161) reported some measurements in
conjunction with the development of an empirical relation for flow double
refraction in pure liquids.

B. Polymers: References 6, 29, 32, 33, 35-37, 39, 42, 49, 50, 74,
75, 79, 91, 93, 95, 116-122, 127, 131, 133, 134, 136, 138, 139, 174, 175,
181, 182, 187, 189, 191, 195, 200, 208, 214, 215, 218-224, 235, 250, 251,
255-257, 268, 270, 271. Much of the recent literature concerning double
refraction of flow has concerned studies on solutions of various polymers.

Many of these polymer molecules are of colloidal dimensions and are so considered by the investigators; however, these are considered with the polymers, rather than the colloids.

Some of the earliest and most extensive work on double refraction of polymer solutions was conducted by Signer and his collaborators (214, 215, 218-224). In 1933 Signer and Gross reported double refraction studies on a large number of nitrocellulose and polystyrene solutions. The effects observed were somewhat similar to those of colloidal systems though not usually as intense as those of such substances as vanadium pentoxide or bentonite. The extinction angle usually approaches 45° at low velocity gradients, but decreases with increasing gradient. The amount of double refraction may vary linearly with velocity gradient or it may increase more or less than linearly, depending upon the molecule under consideration. These phenomena have been studied by Kuhn and Kuhn (131, 133, 134, 136, 138, 139) and are discussed in a review of their work published in 1948 (136).

In 1942 Kanamaru, Tanaka, and Sugiama (117-120) published the results of several investigations on the effects of various factors such as temperature, type of solvent, additives, etc. upon the flow double refraction of polymer solutions. Kuhn and Kuhn, and others, have actually studied the course of polymerization reactions by means of double refraction, and Kuhn and Kuhn (134) developed a quantitative relationship between the degree of polymerization and the amount of double refraction for several systems. Since 1942 Tsvetkov and his collaborators (74, 250, 251, 255-257) considered this application, and conducted a number of double refraction experiments on various polymers, including some work on synthetic and natural rubber solutions.

In 1945 their explanation for some of their results drew criticism from Rehner (185) who showed that certain of their assumptions were inadmissable.

A large number of less extensive investigations of the double refraction of polymer solutions have been made in recent years. These will not be discussed individually here, but references to all of them are listed at the beginning of this section. The review by Cerf and Scheraga (40), both of whom are active in polymer studies, gives an excellent summary of their own work in the field and that of others.

C. Colloids: References 6, 7, 11, 21, 23, 25, 28, 44-47, 50, 57, 58, 66-71, 94, 100, 106, 107, 124, 125, 127, 147, 156, 157, 179, 188, 190, 196, 197, 199, 201, 213, 233, 274, 258, 275, 279. The double refraction effects in colloids are like those in polymer solutions, i. e., not as regular as those in pure liquids. Some of the earliest experimental investigations were made on colloidal substances, and several theories have been developed primarily to explain the colloidal phenomena. Some of the most strongly doubly refracting liquids are colloids, and, for this reason, several have been used in fluid flow studies. (Ref. section on fluid flow analysis.)

From 1915 to 1930 Freundlich, Zocher, and their co-workers (46, 47, 66-70, 279, 280) carried on flow double refraction experiments with several colloids, chiefly the strongly doubly refracting vanadium pentoxide sols. In 1929 Bradfield and Zocher (25) and von Buzagh (28) reported on studies involving sols of bentonite clay. Here, as in vanadium pentoxide, the effect observed is very strong.

Other colloids, while not usually exhibiting the extremely strong double refraction of bentonite and vanadium pentoxide, have been the subject of ex-

tensive studies. In 1925 Kreuger (127) studied a number of colloids along with the pure liquids previously mentioned. Studies on colloidal systems formed a basis for Boeder's (20, 21) theory. Signer (213), in 1930, and Sadron (196, 197) studied a number of colloidal systems and, from them, drew theoretical conclusions. In 1942 Feitknecht, Signer, and Berger (58) made several particle-size determinations on colloidal nickel hydroxide by various methods, including flow double refraction, and obtained excellent agreement between it and the other methods. The double refraction results were calculated from the expressions of Peterlin and Stuart.

A number of other experimental investigations have been made on various colloidal systems. These references will be found listed at the beginning of this section.

D. Proteins: References 10, 12, 22, 32, 36, 37, 43, 50, 51, 54-56, 59-63, 77, 82-84, 86, 94, 98, 102, 105, 110-113, 123, 141-144, 146, 152, 153, 155, 158-160, 164-168, 176, 180, 184, 198, 205, 207, 209-211, 217, 231, 232, 235, 236, 260, 274. A fourth large group of substances studied by flow double refraction methods are the proteins. The initial studies of these substances were described by von Muralt and Edsall. In 1929 they published a preliminary report on their work (158), followed in 1930 by a complete description of their own work, and also a review of previous work in the field of flow double refraction. Edsall and his co-workers, notably Foster (54, 55, 59, 63) and Mehl (56, 152, 153), have reported results on a large number of of protein systems. Scheraga, another of Edsall's collaborators, together with Backus, used flow double refraction to actually follow the clotting of human blood (205) and obtained valuable information as to its mechanism.

Studies by these and other workers have covered size and shape determinations, coagulation phenomena, the effects of heat and additives in denatura-

tion, etc. In 1941 Oncley (168) compared the size and shape values obtained by flow double refraction with those from other methods (including ultra-centrifugation, viscosity, diffusion, etc.). His results were quite reproducible and consistent. Along with his other work, Signer (217) studied proteins and more recently Joly (105, 110-113), Cerf (32, 36, 37), and Schwander (209-211) have studied a number of such systems.

Plant virus constitute an interesting sub-class of proteins studied by flow double refraction. The virus causing tobacco mosaic disease gives intense flow double refraction, thought to be caused by orientation of long rod-shaped particles as in vanadium pentoxide. Studies on tobacco mosaic virus include those of Takahashi and Rawlins (236) in 1937; Lauffer and Stanley (142-144) in 1938-1939; and Kausche, Guggisberg, and Wissler (123) in 1939. Relationships between the molecular properties and the infective power of the virus have resulted from these experiments.

E. Other Substances: A number of other substances beside those mentioned in previous sections have been the subjects of flow double refraction studies. These are listed below along with the references concerning them.

1. Aliphatic Acids and Alcohols (252, 253)

2. Benzopurpurin (14), (15)

3. Carbohydrates (229)

4. Hexadecyltrimethylammonium Bromide (204)

5. Chondroitinsulfuric Acid and Hyaluronic Acid (19)

6. Condensed Phosphates (269)

7. Crystalline Substances p-Azoxyanisole, and Barium Sulfate (246), (249), (261)

8. Liquid Emulsions (170)

9. Milling Yellow Dye (115), (173)

10. Oils (8), (163), (264)

11. Pyroxylin (26)

12. Shellac (192)

13. Soaps and Detergents (2), (5), (9), (76), (80), (81), (95) (114), (226), (242), (243), (280)

III.   Fluid Flow Analysis with Doubly Refracting Liquids

References 1, 9a, 14, 15, 45, 88, 100, 147, 173, 190, 270, 271. The earliest recorded use of a doubly refracting liquid in the analysis of fluid flow was described by Humphry (100) in 1923. He used a vanadium pentoxide sol in his flow apparatus and obtained some simple qualitative results. In 1936 Alcock and Sadron (1), using sesame oil, were able to calculate local velocity gradients from measured values of the double refraction and suggested further applications. Their results were limited by the high viscosity of their test liquid. Hauser and Dewey (88) in 1939 published a short report on experiments then in progress, using bentonite sols, and in 1941 Dewey gave the results in a thesis (45). In this he described a flow facility, presented calibration data and photographs of flow patterns, and showed a method for quantitative calculations of local velocity gradients and streamline directions. The same type of analysis was presented by Weller in 1942 (270, 271). He tested some 50 liquids before finally deciding on a solution of ethyl cellulose (kodak "Ethocel") in a commercial solvent "Methyl Cellosolve" for use in his flow system.

In 1952 Rosenberg (190) presented a summary and extension of the work of Dewey and Weller together with a survey of literature pertaining to flow

double refraction. He also described a flow test apparatus, then under construction, which was designed to utilize a bentonite sol as the double refracting liquid.

In 1945, Leaf (147) described an interesting qualitative application of the doubly refracting liquid to flow analysis. By observing the flow of a bentonite sol inside transparent models of a locomotive fire box, he was successful in determining the flow patterns of the hot gases in the prototype, and in recommending procedures for eliminating erosion of boiler tubes by the ash suspended in these gases. In 1945 and 1947 Binnie (14, 15) described the use of a doubly refracting benzopurpurin sol to observe the onset or turbulence in a long circular tube. In 1947 Ullyott (258) reported the results of some flow experiments using vanadium pentoxide carried out during World War II without knowledge of Dewey's work. Much of Ullyott's work paralleled that of Dewey and Weller, although he seems to have attempted no quantitative analysis of his results. Balint (9a) recently presented a review of a number of methods of flow visualization, including double refraction.

Previous fluid flow investigators have suffered because their doubly refracting liquids have been either extremely viscous or unstable in contact with common materials of construction. In 1953 Peebles, Garber, and Jury (173) reported a new substance for use in fluid flow test facilities, an aqueous solution of a commercial organic dye, milling yellow. Solutions of this dye were found to be stable in contact with common materials of construction, and to give intense flow double refraction in solutions with viscosity little greater than that of water. They described an apparatus then in use for qualitative flow observations, presented photographs of flow patterns, and outlined future quantitative experiments using the dye.

IV. Equipment and Procedures

References 14-16, 18, 26, 34, 40, 50-53, 64, 66, 69, 72, 74, 78, 94, 100, 104, 117, 142, 143, 146, 157, 159, 160, 176, 188, 194, 218, 227, 229, 230, 255, 270-272, 281. A number of the references previously cited, particularly those dealing with experimental measurements, give descriptions of apparatus for measuring flow double refraction. The majority of these are of the concentric cylinder type. (See Edsall (50-54) or Cerf and Scheraga (40) for details of construction on a number of these.) However, a few special types and modifications, as well as several sources dealing with means of eliminating errors in measurement, are given special mention here.

Instead of the concentric cylinders Freundlich (66-70) and Lauffer (143-144) merely allowed their liquids to flow through a narrow tube. However, this method does not permit a definite relationship to be established between the amount of double refraction and the velocity gradient, since the gradient is not constant across the tube. A modification of the concentric cylinder polariscope has been described by Frisman and Tsvetkov (74). It involves three concentric cylinders, instead of the conventional two, with the middle one rotating. This enables tests to be performed with either an inner or an outer rotating cylinder on the same apparatus.

Cerf (34) described an optical method for measuring extinction angles in fluids whose double refraction is too weak to be observed by ordinary methods. Buccheim, Stuart, and Menz (27), and Peterlin and Semac described highly sensitive optical instruments for measuring small effects. Zucker, Foster, and Miller (281) have accomplished similar results using a photoelectric measuring device.

Since no theoretical treatment has been developed for double refraction in turbulent flow, the onset of turbulence in a piece of test apparatus may lead to serious errors. G. I. Taylor (238-242) derived expressions for a critical velocity gradient beyond which turbulence will occur in a concentric cylinder device. Jerrard (104) surveyed the apparatus of some 30 investigators and presented a convenient tabulation of the results of Taylor's work so that the critical gradient for any concentric cylinder polariscope may be easily calculated from its dimensions and the viscosity of the liquid being tested. Turbulence and other sources of error have been considered by Björnstahl (16, 19), Frey-Wissling and Weber (72), and Snellman (237). These writers describe conditions under which given sources of error may arise along with methods of eliminating them.

V. Background Material

In order to provide information on the underlying principles of flow double refraction, some selected references in related fields are included in this summary. These are listed below grouped according to their content.

A. General Optical Theory: References 41, 45, 190, 267.

B. Principles of Double Refraction (non-flow): References 41, 45, 190, 229, 267, 272.

C. Flow of Fluids between Concentric Cylinders: References 104, 128, 129, 171, 238, 240, 241, 261.

D. Relation of Molecular Form to Viscosity and Flow Behavior: References 26, 30, 33, 65, 81, 99, 101, 103, 120, 135-137, 141, 145, 187, 188, 239, 261.

E. Electrical and Magnetic Double Refraction: References 169, 233, 237, 247-249.

F. **Dielectric Measurements**: Reference 157.

VI. **Reviews**

Articles reviewing the field of flow double refraction are listed in the Bibliography as follows: 40, 49, 50, 51, 94, 96, 117, 133, 136, 159, 177, 190, 230.

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APPENDIX B

NOMENCLATURE

Symbol	Meaning
a	Amplitude of light wave transmitted by the polarizer
c	Velocity of light in air
c_e	Velocity of light vibrating parallel to the optic axis
c_o	Velocity of light vibrating normal to the optic axis
E_s	Modulus of elasticity in shear
f	Functional designation
g	Functional designation
K	Instrument constant for the MacMichael viscometer
L	Capillary length
L_w	Length of wire supporting the inner cylinder of the MacMichael viscometer
n_e	Refractive index of light vibrating parallel to the optic axis
n_o	Refractive index of light vibrating normal to the optic axis
N	Integer designation, or fringe order number
ΔP	Pressure drop across the capillary viscometer
Q	Volumetric flow rate
r	Radial distance
r_i	Radius of the inner cylinder of a concentric cylinder flow cell
r_2	Radius of the outer cylinder of a concentric cylinder flow cell
r_w	Radius of the wire supporting the inner cylinder of the MacMichael viscometer
R	Capillary tube radius

Symbol	Meaning
s	Liquid depth
t	Time
u	Vibration displacement of light transmitted by the polarizer
u_1	Vibration displacement, normal to the optic axis, of light entering the doubly refracting liquid
u_2	Vibration displacement, parallel to the optic axis, of light entering the doubly refracting liquid
u_1'	Vibration displacement, parallel to the optic axis, of light transmitted by the doubly refracting liquid
u_2'	Vibration displacement, normal to the optic axis, of light transmitted by the doubly refracting liquid
u_1'	Vibration displacement, parallel to OX, of light entering the quarter wave plate
u_2'	Vibration displacement, parallel to OY, of light entering the quarter wave plate
u_1''	Vibration displacement, parallel to OX, of light transmitted by the quarter wave plate
u_2''	Vibration displacement, parallel to OY, of light transmitted by the quarter wave plate
v	Liquid velocity
w	Vibration displacement of light transmitted by the analyzer with the plane of polarization parallel to OY
w'	Vibration displacement of light transmitted by the analyzer with the plane of polarization parallel to OY'

Symbol	Meaning
x	Arbitrary phase retardation
α	Arbitrary angle
δ	Retardation distance produced by the doubly refracting liquid
Θ	Angle necessary to rotate the analyzer to measure double refraction using the quarter wave plate compensator
Θ_t	Angular displacement of the inner cylinder of the MacMichael viscometer
λ	Wave length of light used in the concentric cylinder polariscope
μ	Liquid viscosity
μ_a	Apparent liquid viscosity
π	Ratio of circumference to the diameter of a circle
τ	Local shear stress in a moving liquid
τ_w	Wall shear stress
ϕ	Angle between the normal to the optic axis and the plane of polarization of the polarizer
χ	Extinction angle
ω	Angular velocity of the outer cylinder of a concentric cylinder flow cell

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Department of Mechanical
Engineering
Johns Hopkins University
Baltimore 18, Maryland (1)

Dr. L. G. Straub
St. Anthony Falls Hydraulic
Laboratory
University of Minnesota
Minneapolis 14, Minnesota (1)

Dr. G. H. Hickox
Engineering Experiment
Station
University of Tennessee
Knoxville, Tennessee (1)

Director
Woods Hole Oceanographic
Institute
Woods Hole, Massachusetts (1)

Mr. C. A. Gongwer
Aerojet General Corporation
6352 N. Irwindale Avenue
Azusa, California (1)

Stevens Institute of Technology
Experimental Towing Tank
711 Hudson Street
Hoboken, New Jersey (1)

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Department of the Navy
Washington 25, D. C.
Attn: Code Re6a   (1)

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Department of the Navy
Washington 25, D. C.
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National Hydraulic Laboratory
National Bureau of Standards
Washington 25, D. C.   (1)

Professor H. Reissner
Department of Aeronautical
      Engineering and
      Applied Mechanics
Polytechnic Institute of
      Brooklyn
99 Livingston Street
Brooklyn 2, New York   (1)

Brown University
Graduate Division of Applied
      Mathematics
Providence 12, Rhode Island   (1)

California Institute of Tech-
      nology
Hydrodynamics Laboratory
Pasadena 4, California
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Professor G. Birkhoff
Department of Mathematics
Harvard University
Cambridge 38, Massachusetts   (1)

Professor G. F. Wislicenus
Department of Mechanical
      Engineering
Johns Hopkins University
Baltimore 18, Maryland   (1)

Massachusetts Institute of
      Technology
Department of Naval Archi-
      tecture
Cambridge 39, Massachusetts   (1)

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State College, Pennsylvania   (1)

Stanford University
Applied Mathematics and
      Statistics Laboratory
Stanford, California   (1)

Professor H. A. Einstein
Department of Engineering
University of California
Berkeley 4, California   (1)

Professor C. H. Wu
Department of Mechanical
     Engineering
Polytechnic Institute of Brooklyn
99 Livingston Street
Brooklyn 2, New York   (1)

Professor V. L. Streeter
Civil Engineering Department
University of Michigan
Ann Arbor, Michigan   (1)

Commander
Naval Ordnance Test Station
3202 E. Foothill Blvd.
Pasadena, California   (1)

Commanding Officer and Director
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Oak Ridge, Tennessee
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      Dr. R. N. Lyon   (1)

Dr. Thomas Baron
Shell Development Company
Emoryville, California   (1)

A. J. Gram
Research Center
The Babcock and Wilcox Company
P. O. Box 535
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R. D. Brooks
Knolls Atomic Power Laboratory
Peek Street Site
P. O. Box 1072
Schenectady 1, New York   (1)

Dean K. E. Schoenherr
College of Engineering
University of Notre Dame
Notre Dame, Indiana   (1)

Professor J. L. Hooper
Alden Hydraulic Laboratory
Worcester Polytechnic
     Institute
Worcester, Massachusetts   (1)

Chief, Bureau of Ships
Department of the Navy
Washington 25, D. C.
Attn: Research Division   (1)
      Code 420 (Preliminary
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Hydrodynamic Laboratory
Pasadena 4, California   (1)

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Assistant Director, Engineer-
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Engineering Department
E. I. DuPont De Nemours and
     Company, Inc.
Wilmington 98, Delaware   (1)

University Laboratory of
     Physical Chemistry
Harvard University
25 Shattuck Street
Boston 15, Massachusetts
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      Dr. Barbara W. Low   (1)

Dr. C. E. Brehm, President
University of Tennessee
Knoxville 16, Tennessee   (1)

Dr. E. A. Waters, Vice
     President
University of Tennessee
Knoxville 16, Tennessee   (1)

W. B. Giles
General Electric Company
One River Road
Schenectady 5, New York   (1)

Dr. Leland G. Cole, Head
Rocket Propulsion Laboratory
University of Michigan
Willow Run Airport
Ypsilanti, Michigan   (1)

Dr. Frank A. Anderson
Department of Chemistry and
          Chemical Engineering
University of Mississippi
University, Mississippi   (1)

The Franklin Institute
Philadelphia 3, Pennsylvania
Attn: A. O. Bengholm
          Engineering Mechanics
          Section   (1)

A. L. Pollard
514 21st Street
Knoxville, Tennessee   (1)

Department of Chemical Engineering
University of Tennessee
Knoxville 16, Tennessee
Attn: Dr. R. M. Boarts   (1)
      Professor H. J. Garber   (1)
      Dr. E. E. Stansbury   (1)
      Dr. H. F. Johnson   (1)
      Dr. S. H. Jury   (1)
      Dr. A. deS. Brasunas   (1)

Dean N. W. Dougherty
College of Engineering
University of Tennessee
Knoxville 16, Tennessee   (1)

University of Tennessee
Department of Chemistry
Knoxville 16, Tennessee
Attn: Dr. C. A. Buehler   (1)
          Dr. H. A. Smith   (1)

Dr. J. F. Bailey
Department of Mechanical
          Engineering
University of Tennessee
Knoxville 16, Tennessee   (1)

Professor A. T. Granger
Department of Civil
          Engineering
University of Tennessee
Knoxville 16, Tennessee   (1)

Dr. K. L. Hertel
Department of Physics
University of Tennessee
Knoxville 16, Tennessee   (1)

Dr. W. B. Harrison
School of Mechanical
          Engineering
Georgia Institute of Tech-
          nology
Atlanta, Georgia   (1)

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